AN INVESTIGATION INTO THE USE OF BIOKINETIC MODELS WHEN ASSESSING INTAKES OF PLUTONIUM

A Thesis

by

BRIAN ANDREW HRYCUSHKO

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

August 2008

Major Subject: Health Physics

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Approved by:

Chair of Committee,	John W. Poston Sr.
Committee Members,	Leslie A. Braby
	Michael Walker
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ABSTRACT

An Investigation into the Use of Biokinetic Models When Assessing Intakes of Plutonium. (August 2008)

Brian Andrew Hrycushko, B.S.; B.S., Texas A&M University

Chair of Advisory Committee: Dr. John W. Poston Sr.

The goal of internal dosimetry is to assess the dose to an individual from an intake of a radionuclide. This usually encompasses assessing the intake amount based on some form of bioassay measurement used with a biokinetic model. There are many published biokinetic models that describe the transfer of radionuclides throughout the body. It would be beneficial at times if one could interchange certain biokinetic models with another to assess an intake based on bioassay data to save time and make calculations simpler. This research compared the daily excretion rates by interchanging widely used biokinetic models in different combinations. These model combinations were then used to assess an unknown intake of a case study.

It was shown that the ICRP-30 and ICRP-66 respiratory tract models can only be interchanged at specific times post intake to give similar excretion results from an inhalation intake. It is feasible to interchange the ICRP-67 plutonium systemic model or the newer Luciani and Polig plutonium systemic model to assess an intake based on fecal bioassay data, but not urine bioassay data for ingestion intakes. It is not feasible to interchange the systemic models when assessing intakes from a wound or injection.

Using different combinations of biokinetic models predicted intakes within 30% for a case study which included a relatively long inhalation chronic intake followed by a much shorter chronic inhalation intake. It was shown that the predicted initial chronic intake for each combination of models gave fecal excretion values which deviated the most from the worker's fecal bioassay data. This could mean that the biokinetic models yield inaccurate excretion rates for long chronic intakes.

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Finally, thanks to my friends at Fairview. Who would have thought we would be where we are now?

NOMENCLATURE

Bq	Becquerel
d	Day
g	Gram
Gy	Gray
kg	Kilogram
MeV	Megaelectron volt
mSv	Millisievert
Pu	Plutonium
s	Seconds
Sv	Sievert
μm	Micrometer

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CHAPTER I

INTRODUCTION

In order to interpret the measurement of activity in excreta it is important to have knowledge of the behavior of radioactive materials within the human body. The International Commission on Radiological Protection (ICRP) has developed several biokinetic models over the years to describe the transfer of activity throughout the body and through routes of excretion. Some of the models most often used are described in ICRP Publications 23 (ICRP 1975), 30 (ICRP 1979), 56 (ICRP 1989), 66 (ICRP 1994), 67 (ICRP 1993), and 78 (ICRP 1997). These models are based on linear, first-order kinetics with parameters that can be modified so that radionuclide flow throughout the body can be represented. Biokinetic models have been used for dose calculations, setting dose limits, and for assessing intakes of radiation based on bioassay measurements. Fig. 1 shows the general form of a biokinetic model of the human body (ICRP 1997). Arrows indicate radionuclide transfer, and boxes indicate specific compartments within the body where radionuclide uptake occurs. Published ICRP models describe the respiratory tract, gastrointestinal tract, and systemic systems in more detail.

This thesis follows the style of Health Physics.

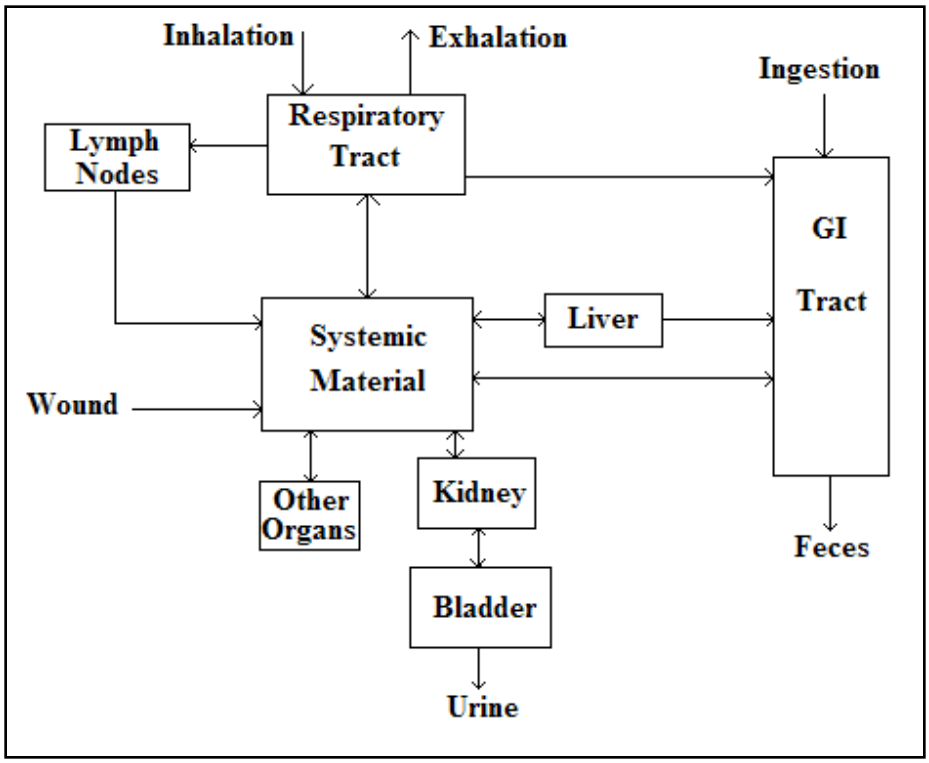


Fig. 1: General compartmentalized biokinetic model of the human body (Adapted from ICRP 1997)

It is desired to use bioassay measurements with biokinetic models when assessing intakes of radionuclides. The bioassay measurements used to assess intakes of radionuclides can include: *in vivo* measurements, excretion measurements, air monitoring measurements, etc. The order of preference of assay type, in terms of ease of measurements and importance of interpretations is body activity analysis, excretion analysis, followed by personal air sampling (ICRP 1997). Body activity analysis *in vivo* can be used to directly assess the amount of activity in the body, but may only be useful for radionuclides that emit x or γ radiation. Excreta measurements can be used to assess intakes, but there is error due to large differences in the rates at which radionuclides traverse the body and fluctuations in the amount of an individual's daily excretion. The

interpretation of air samples may be misleading as the location of the air monitors can affect measurements by a factor of about ten (ICRP 1997). Many bioassay programs rely on the analysis of excretion samples as part of their internal exposure control program due to lower cost and ease of collection. It would be beneficial to be able to use the excretion measurements with simple biokinetic models to assess correct radionuclide intakes with little error. This would ultimately cut both time and costs of other more accurate, but unreasonable intake assessment methods; however, it can often be computationally arduous using more complicated models to assess intakes. Interchanging models or subsystems of models may speed up the assessment process while still providing valid results.

Urinary excretion is removal of material from the body in urine through the bladder. Fecal excretion is the removal of material that either passes unabsorbed through the gastrointestinal tract (GI-tract) or is comprised of systemic material that is removed through the GI-tract. For some radionuclides, including plutonium, routes of excretion are given explicitly in biokinetic models (ICRP 1993, Luciani and Polig 2000), while other models split excreted activity between urine and feces depending on a constant ratio (ICRP 1989). It can be a difficult task to match the excretion predictions from models that split total excretion to actual bioassay measurements, especially for chronic intakes over long periods of time or cases where radionuclide intake amounts fluctuate over time.

Plutonium intakes present a problem because it is difficult to measure and interpret bioassay data. Monitoring programs usually collect urine samples due to the

ease of collection and esthetic reasons (Cember 1996); however, fecal analysis is preferred for such radionuclides such as plutonium with a clearance class of Y (ICRP 1979) or S (ICRP 1994). This is because the levels of this class of material in excretion can be several orders of magnitude higher in feces than in urine days after a single inhalation intake (Khokhryakov et al. 2004). Urine bioassay data may be more feasible for other intake modes, such as wounds or injections, where urine activity levels are the same as that of fecal activity levels.

OBJECTIVES

The objective of this research is to compare excretion predictions from interchanging current and past biokinetic models and use these models to assess an unknown intake of plutonium using excretion data. This is important because plutonium is one of the most widely seen elements in occupational exposures. The goal in assessing an unknown intake is to distinguish the correct mode of intake, the amount of radionuclide intake, and the time frame of the intake. This will be accomplished by developing popular published biokinetic models using the computer program Simulink to represent radionuclide transfer through the body and analyze excretion data. Specific goals include:

- Investigate differences in predicted daily excretion for an acute inhalation intake of ^{239}Pu when interchanging the ICRP Publication 30 (ICRP-30) respiratory tract model and the ICRP Publication 66 (ICRP-66) respiratory tract models with the ICRP Publication 67 (ICRP-67) and the Luciani and Polig plutonium systemic models. This investigation will consider the possibility of interchanging the

respiratory models when assessing an intake from excretion data due to the difficulty in implementing the ICRP-66 respiratory tract model and all its detail. Interchanging the ICRP-30 respiratory tract model for the ICRP-66 respiratory tract model could speed up intake assessment and still provide valid results.

- Investigate differences in plutonium excretion for acute wound/injection and ingestion intakes of ^{239}Pu using the ICRP-67 systemic model and a more recent plutonium systemic model by Luciani and Polig (Luciani and Polig 2000). This is done because the Luciani and Polig systemic model has been known to give better agreement with measured bioassay excretion and higher estimates of intake compared with current ICRP models. This research will investigate the possibility of interchanging the biokinetic models when assessing intakes from excretion data.
- Use the biokinetic models in a practical application to assess the intake for a case study where there was an unknown intake of ^{239}Pu by a worker. This is done to compare intake assessments and dose calculations based on excretion measurements for ^{239}Pu using different combinations of interchanging the biokinetic models.

CASE STUDY OF UNKNOWN INTAKE

The biokinetic models will be used to assess the intake for a case study. A worker received a leg laceration during mixed waste processing operations. Direct survey of the workers clothing indicated the presence of contamination, but no contamination was found at the wound site. Twenty-four-hour urinary excretion data

were collected periodically and ^{239}Pu content was determined. However, initial measurements suggested a different mode of intake than that from a wound. Single-event fecal samples and twenty-four-hour urine samples were then taken periodically over a course of several months. After the laceration, the worker was placed on work restriction in areas that did not include elevated radioactivity levels to prevent additional intakes. This worker had been employed at the waste processing site for about 22 months prior to the laceration event, but did not work in any radiological areas for a period of over two months in the middle of employment. Area monitoring showed relatively constant low levels of airborne activity and did not indicate elevated levels of activity. About two months after the worker was placed on restriction, air monitors showed a brief five-day spike in airborne activity in non-radioactive work areas that could indicate a second intake. There were a total of seven fecal measurements with six positive for ^{239}Pu. There were a total of seven urinary measurements with only one positive for ^{239}Pu. The one positive urine sample was at such a low level that only fecal measurements are used with the biokinetic models in this study. The combinations of biokinetic models will be used to assess the mode of radionuclide intake, the amount of radionuclide intake, and the time frame of the radionuclide intake.

CHAPTER II

THEORY

ICRP PUBLICATION 30 RESPIRATORY TRACT MODEL

ICRP Publication 2 (ICRP 1959) served as an adequate criterion for the controlling of internal radiation exposure, but the maximum permissible concentration in air (MPC) and the maximum permissible body burden (MPBB) were often misused or misunderstood. The ICRP-30 respiratory model was an improvement from the ICRP Publication 2 respiratory model. New information on the relationship between ionizing radiation and risks to biological effects in the body was the basis for establishing limits for exposure to ionizing radiation (ICRP 1979). The ICRP-30 respiratory model is based on a design proposed by the Task Group on Lung Dynamics for Committee II (Task Group 2) of the International Radiological Protection Commission. The Task Group 2 design determines deposition in and clearance from the human respiratory tract in order to calculate dose to the lung and create standard exposure limits. The committee compartmentalized the lung to differentiate deposition and clearance mechanisms (ICRP 1979). The compartmentalization depended on lung anatomy, particle deposition characteristics, and particle clearance characteristics.

The ICRP-30 respiratory model is separated in to three regions: the nasal passage or the naso-pharyngeal region (N-P), the trachea and bronchial region (T-B), and the pulmonary parenchyma region (P). The N-P region is the upper region of the lung starting at the anterior nares and continues down to the larynx. The T-B region consists of the trachea and bronchial tree down through the terminal bronchioles. This region,

together with the N-P region, contains the entire epithelial area of the respiratory tract that is ciliated and covered with mucus coming from columnar epithelial cells and secretory glands. The P, together with the T-B region, is the lower respiratory tract. The P region consists of the respiratory bronchioles, the alveolar ducts, and the alveoli. The surface of the P region is made up of epithelial cells that are not ciliated.

Deposition is assumed to depend on the aerodynamic properties of the aerosol in question and is determined by the activity median aerodynamic diameter (AMAD). The AMAD is the diameter of a unit sphere that has the same settling velocity as the particle (ICRP 1979). The ICRP-30 respiratory model is meant to be used for particles ranging from 0.2 μm to 10 μm AMAD. The parameters D_{N-P}, D_{T-B}, and D_P represent the fraction of inhaled material deposited in each region of the lung. These fractions are given on page 25 in ICRP-30 for different AMAD sizes (ICRP 1979). If the size of inhaled particles is unknown, a 1μm AMAD standard sized particle is used where: D_{N-P} is 0.3, D_{T-B} is 0.08, and D_P is 0.25.

Particle clearance has been investigated in many studies involving animals and humans. When particles are deposited in the lungs they undergo clearance mainly by endocytosis or ciliary movement of mucus. Endocytosis clearance depends on the number, size, shape, and surface reactivity of the particles while clearance by ciliary movement seems to be relatively constant. Each of the three regions of the lung is separated into more compartments based on different modes of particles clearance. The ICRP-30 respiratory model considered all published quantitative clearance information at the time to assess particle clearance rates. Three classifications of clearance rate

describe removal from the lung: class D materials have clearance half times of less than ten days, class W materials have clearance half times from ten to one hundred days, and class Y materials have half times greater than one hundred days. Clearance rates from compartment to compartment are defined based on the class of particle. The activity q (Bq) in compartment i at time t (days) after intake is calculated using eqn (1) (Khokhryakov et al. 2005).

$$\frac{dq_i(t)}{dt} = \dot{I}(t)D_iF_i + \sum_{\substack{j=1 \\ j\neq i}}^{n} \lambda_{j,i}q_j - (\lambda_R + \sum_{\substack{j=1 \\ j\neq i}}^{n} \lambda_{i,j})q_i \quad (1)$$

where $\dot{I}$ (t) is the radionuclide intake rate (Bq/day) at time t by inhalation, D_i is the fraction of the inhaled activity deposited in compartment i, $\lambda_{j,i}$ is the transfer coefficient from compartment j to compartment i, $\lambda_{i,j}$ is the transfer coefficient from compartment i to compartment j, λ_R is the radioactive decay constant, and n is the number of compartments used in the model. This equation describes radionuclide flow through the respiratory tract through linear, first-order kinetics. D_i is zero for the lymph node compartments. Fig. 2 shows the compartmentalized ICRP-30 respiratory model. Arrows define a clearance pathway for radionuclide flow. Compartments a, c, and e represent absorption processes into the systemic circulation. Compartments b, d, f, and g represent particle transport processes to the GI-tract (ICRP 1979). Compartment h represents the slow removal from the pulmonary region to the lymphatic system. Material sent to compartment i is transferred to the blood, while material sent to compartment j is permanently retained. The ICRP-30 respiratory model was mainly

used in combination with the other ICRP-30 dosimetry models (gastrointestinal tract, bone model, systemic model) to establish dose limits and calculate doses to the body or organs from known intakes. It is not used to assess intakes using bioassay measurements as there are no specific excretion pathways (Task Group 1966, ICRP 1979).

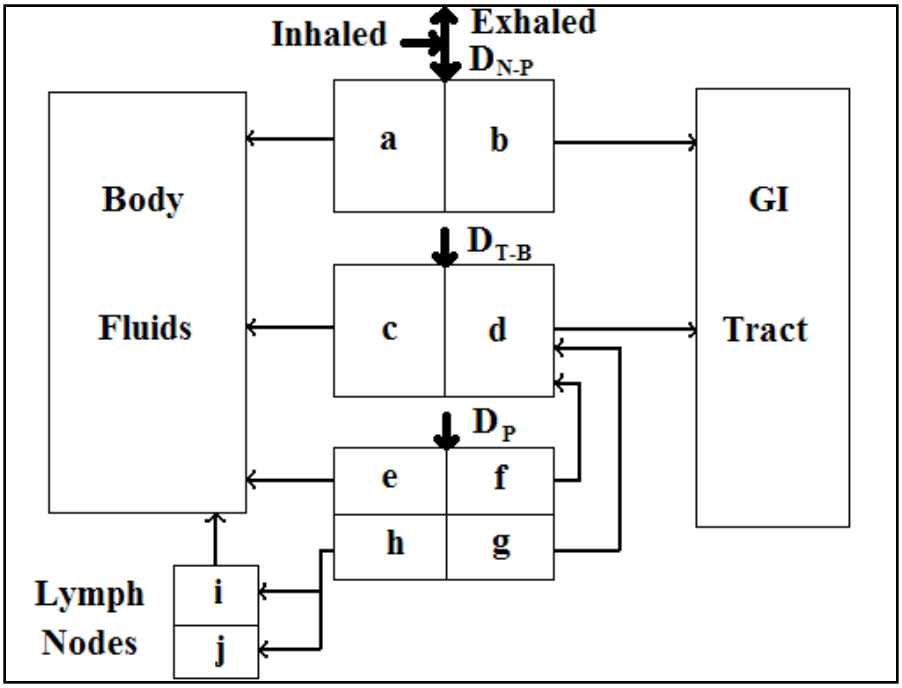


Fig. 2: ICRP Publication 30 respiratory tract model
(Adapted from ICRP 1979)

ICRP PUBLICATION 66 RESPIRATORY TRACT MODEL

A task group was created to review the respiratory model presented in ICRP-30 and make revisions based on research since the 1960's. This was done in part because many studies involving animals resulted in radioactive material being cleared from the respiratory tract at different rates than those given by the ICRP-30 model. The D, W, and Y classifications were revised. The research used to derive the model entailed

greater knowledge of the anatomy and physiology of the respiratory tract, the deposition and clearance of particles, and the biological effects of radioactive particles. The ICRP-30 respiratory model is used to calculate dose by averaging over the entire mass of the lung while the ICRP-66 respiratory model takes into account differences in radiation sensitivity among the different tissues of the lung and allows the calculation of dose to these specific tissues. This respiratory model is much more complex than that of the ICRP-30 respiratory model as more detail is taken into consideration including: age, race, sex, breathing characteristics and habits, health detriment of the lungs, and target tissues with different sensitivities to radiation (ICRP 1994). The model of the ICRP-66 respiratory tract considers the morphometry of the respiratory tract, the respiratory physiology, the radiation biology, deposition, clearance, and dosimetry.

The morphometry of the respiratory tract describes structure and dimensions useful in calculating doses. The respiratory tract is represented as four separate anatomical regions: the extra thoracic region (ET), the bronchial region (BB), the bronchiolar region (bb), and the alveolar-interstitial region (AI). Separation of the regions is based on anatomical and physiological characteristics along with radiobiological response (ICRP 1994). The ET region is composed of the ET_1 (anterior nose) and the ET_2 (posterior nasal passages, larynx, pharynx, and mouth). This is the N-P region of the ICRP-30 respiratory model. The BB region is composed of the trachea and bronchi while the bb region is made up of the bronchioles and terminal bronchioles. The BB and bb regions together are the T-B region of the ICRP-30 respiratory tract model. The AI region, which is composed of the respiratory bronchioles, alveolar ducts,

and the interstitial connective tissue, is the P region of the ICRP-30 respiratory tract model. Each region contains lymphatic tissue with the ET regions initially clearing to the extra-thoracic lymph nodes (LN_{ET}) and the BB, bb, and AI regions clearing to the thoracic lymph nodes (LN_{TH}). Average values of cell depths in each region of the lung are interpreted from limited information and can be found in ICRP-66 (ICRP 1994). The main advantage of the use of the ICRP-66 respiratory tract model is the ability to tailor parameters specifically to individuals.

A deposition model was created to determine the fraction of inhaled material that gets deposited to each region of the respiratory tract (Bair 1995). ICRP-66 tabulates deposition fractions for several sized particles. Each region of the respiratory tract acts as a filter with a filtering efficiency and deposition depending on:

1. the activity mean thermodynamic diameter (AMTD), which determines deposition by thermodynamic processes
2. the AMAD, which determines deposition by impaction and settling.

The rate at which material clears each compartment depends on the particle transport processes that move material to the GI-tract and lymph nodes and the absorption processes moving material to the blood. The bronchial and bronchiolar regions have varying phases of clearance. To account for the varying phases, these regions were each divided in to three compartments: normal phase clearance, slow phase clearance, and a separate compartment where activity is transferred to the lymph nodes over time (sequestered compartment) (Bair 1995). There are also three separate phases of clearance seen in the alveolar region and it, too, is divided into three compartments. The

absorption process entails both dissolution and uptake. Dissolution is when particles separate to materials that can be absorbed by the blood and uptake is when the material dissolved from particles gets absorbed to the blood. To estimate a time-dependent transfer rate, a fraction (f_r) dissolves fast (at a rate s_r), and a fraction ($1-f_r$) dissolves slower (at a rate s_s). To estimate a time-dependent transfer rate which does not have to decrease with time, deposited material simultaneously dissolves to blood (at a rate s_p) and to a transformed state (at a rate s_{pt}), where is dissolves to blood (at a rate s_t). Uptake to blood can be slower due to material binding in the respiratory tract. Fig. 3 shows the compartmental model for the ICRP-66 respiratory tract. Compartments 1-3 and 14-16 are the AI_1, AI_2, and AI_3 compartments, respectively. Compartments 4-6 and 17-19 are the bb_1, bb_2, and bb_{seq} compartments, respectively. Compartments 7-9 and 20-22 are the BB_1, BB_2, and BB_{seq} compartments, respectively. Compartments 10 and 23 are LN_{TH} compartments. Compartments 11, 12, 24, and 25 are the ET_2 compartments, and compartments 13 and 26 are the LN_{ET} compartments. The differential equations describing the flow of radionuclides from compartment to compartment are described by eqn (1) (Khokhryakov et al. 2005).

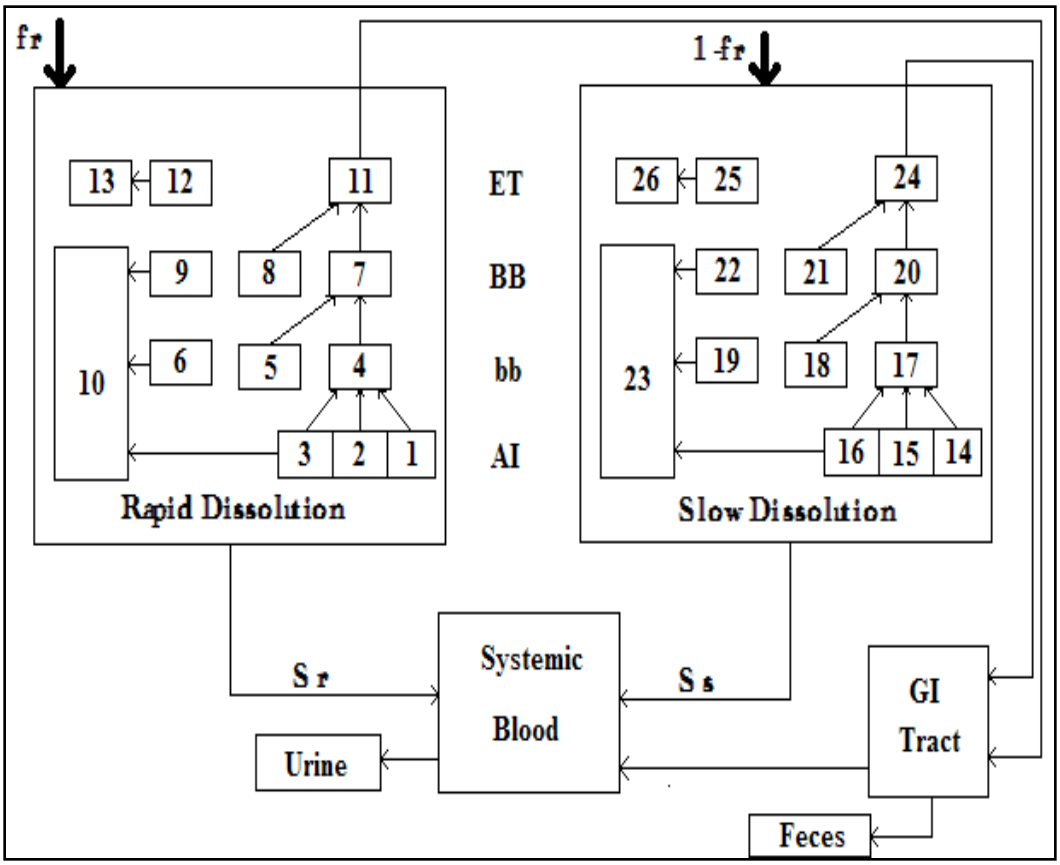


Fig. 3: Compartmentalized model of the ICRP Publication 66 respiratory tract (Adapted from ICRP 1994)

ICRP PUBLICATION 30 GASTROINTESTINAL TRACT MODEL

It is difficult to model the GI-tract because physiological parameters vary from person to person. Physiological parameters also vary within an individual depending on eating habits and the health of the individual (Skrable et al. 1975). The dosimetric model of the ICRP-30 GI-tract is based on the model developed by Eve (Eve 1966). The ICRP-30 GI-tract model is made up of four compartments: the stomach, the small intestine (SI), the upper large intestine (ULI), and the lower large intestine (LLI). This model treats the GI-tract as a tube through which food will constantly flow until it is excreted at

the end (ICRP 1979). Although absorption of some of the gut contents takes place in the SI, ULI, and LLI, it is assumed that radioactive material can only be absorbed to the blood and to the rest of the body through the SI compartment. The fraction of activity transferred to the blood is controlled by the F_1 value designated to each element and implies that the amount of activity reaching the ULI and LLI compartments depends on F_1 (Dolphin and Eve 1966). The transfer rates from compartment to compartment and the F_1 values for different radionuclides can be found in ICRP-30 (ICRP 1979). These values are based on human studies (Eve 1966). Eqn (2) shows the form of the differential equations used to describe the transfer of activity through the GI-tract. $\dot{I}(t)$ is the rate of ingestion of activity of the radionuclide at time t. This can come from the respiratory tract in the case of an inhalation, from an intake occurring by ingestion, or from the blood in the case of a wound or injection.

$$\frac{dq_i(t)}{dt} = \dot{I}(t) + \sum_{\substack{j=1 \\ j \neq i}}^{n} \lambda_{j,i} q_j - (\lambda_R + \sum_{\substack{j=1 \\ j \neq i}}^{n} \lambda_{i,j}) q_i \qquad (2)$$

Each compartment of the GI-tract is made up of the contents and the wall. The contents are considered the source of the radiation and the walls are considered the target of the radiation (ICRP 1979). Epithelium cells interfering with the contents of GI-tract organs are not of importance because they are discarded and damage received by these cells is not retained in the sensitive tissue (Eve 1966). The sensitive cells of the epithelium are located on the mucosal surface of the GI tract and are formed deep in the

crypts of the mucosa. These cells are then pushed up to the contents after a few days (ICRP 1979). The distance between the contents and the sensitive cells causes for the use of absorbed fractions that are different depending on radiation penetrability. This is because not all of the energy of the particle is absorbed in the target. Fig. 4 shows the compartmental ICRP-30 GI-tract model.

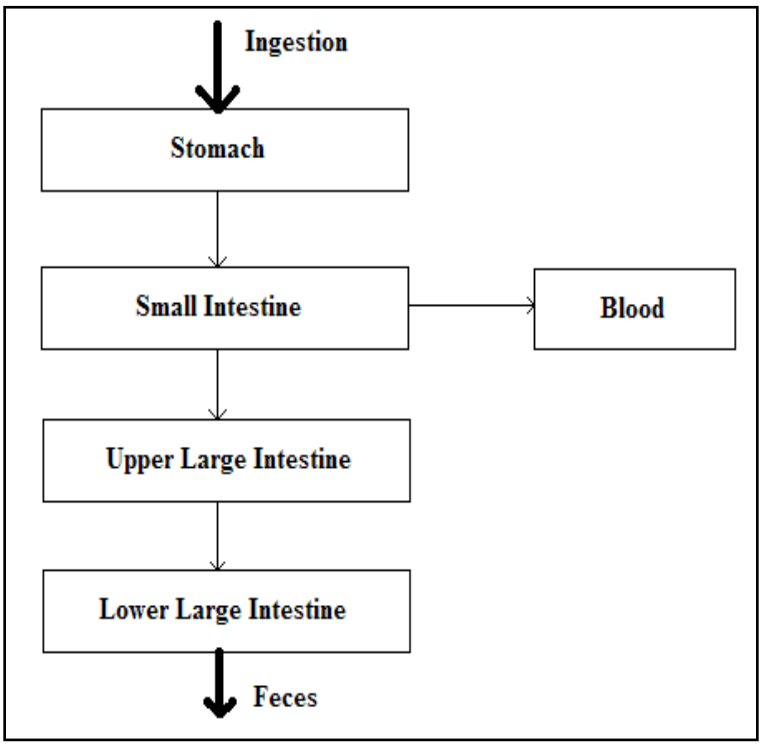


Fig. 4: Compartmentalized model of the ICRP Publication 30 GI-tract model (Adapted from ICRP 1979)

ICRP PUBLICATION 67 PLUTONIUM SYSTEMIC MODEL

For some radionuclides, as is the case for plutonium, there are specific systemic biokinetic models used to describe activity transfer. The old method for determining an intake of plutonium given in ICRP Publication 56 (ICRP 1989) was modified based on

new information of greater retention in soft tissues (ICRP 1997). Also, the previous models of plutonium did not include the specific excretion pathways for plutonium leaving the body. The ICRP-67 systemic model of plutonium considers the uptake, retention, and excretion of activity in greater detail than past systemic models. This model is based on the age-specific biokinetic model for americium (Leggett 1992), with the exception that there are two liver compartments for plutonium. Transfer rates are based on *in vivo*, excretion, and autopsy measurements (ICRP 1993). Activity is transferred to the blood compartment in the systemic model from the respiratory tract and the GI-tract. The ST0 compartment is considered soft tissue comprised of extra-cellular fluids. It exchanges activity with the blood in a short period of time (hours or days). Compartments ST1 and ST2 are used to represent large soft tissue (muscle, skin, etc.) with a longer period of exchange with the blood (years). The activity transferred to the skeleton is divided among the cortical and trabecular bone. Each of these is subdivided further into volume, surface, and marrow compartments. Radionuclides initially get deposited on the bone surfaces, transferring to the bone marrow by resorption and to the bone volume during bone formation. Urine excretion from the urinary bladder contents may occur at a rate of 12 d^{-1}. In addition to unabsorbed ingesta, feces from the GI-tract contain bile from the liver and secretions from the blood (ICRP 1993). Fig. 5 shows the compartmental ICRP-67 plutonium systemic model.

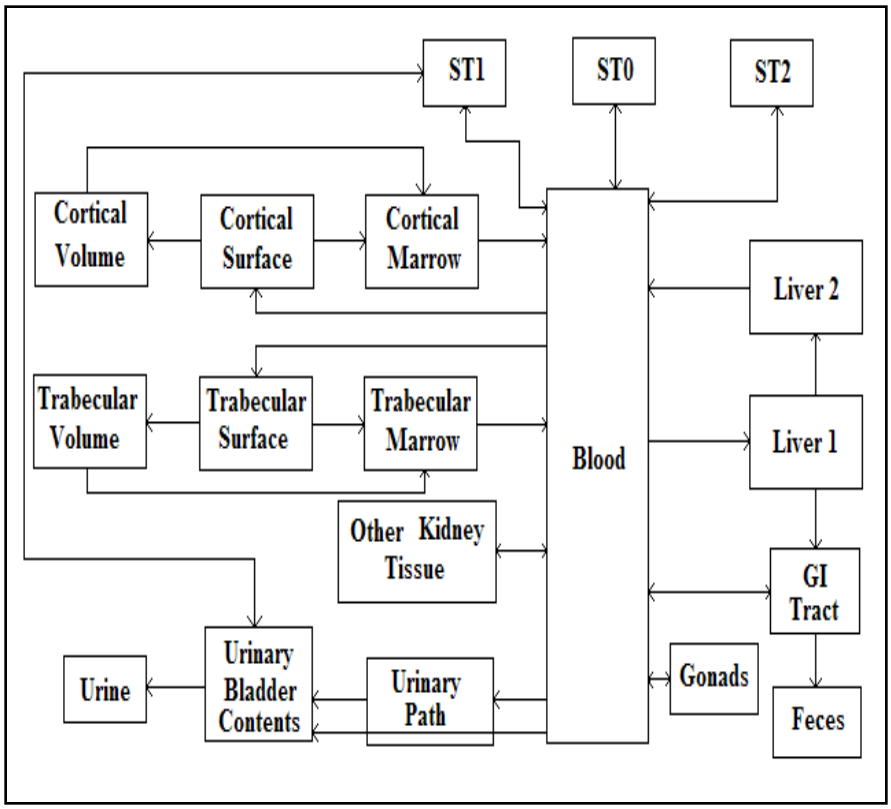


Fig. 5: Compartmentalized ICRP Publication 67 plutonium systemic model (Adapted from ICRP 1993)

LUCIANI AND POLIG PLUTONIUM SYSTEMIC MODEL

The excretion results using the ICRP-67 plutonium systemic model do not quite fit with data from experimental studies and occupational exposures for short time periods after exposure (weeks) for injection studies (Luciani and Polig 2000). The ICRP-67 model assumes that radionuclides are also transferred from the ST1 soft tissue compartment to the urinary bladder compartment. The majority of the radionuclide excreted through urine is reportedly transferred through this route, but does not make sense physiologically (Luciani and Polig 2000). The Luciani and Polig systemic model shows much better agreement with experimental studies and occupational exposures

including the curve constructed by Khokhryakov et al. (Khokhryakov et al. 2004), which was based on a large database of excretion data. The Luciani and Polig plutonium systemic model is the same as the ICRP-67 systemic model for plutonium, with some exceptions. The activity that was transferred from the ST1 compartment to the urinary bladder compartment now goes to the blood compartment. Some of the transfer rates from compartment to compartment were changed based on studies on blood activity, autopsy data, fecal excretion, and urinary excretion optimization (Luciani and Polig 2000). Also, the bone system was altered to allow for time-dependent transfer rates, as suggested in ICRP Publication 70 (ICRP 1995). Fig. 6 shows the compartmentalized Luciani and Polig plutonium systemic model.

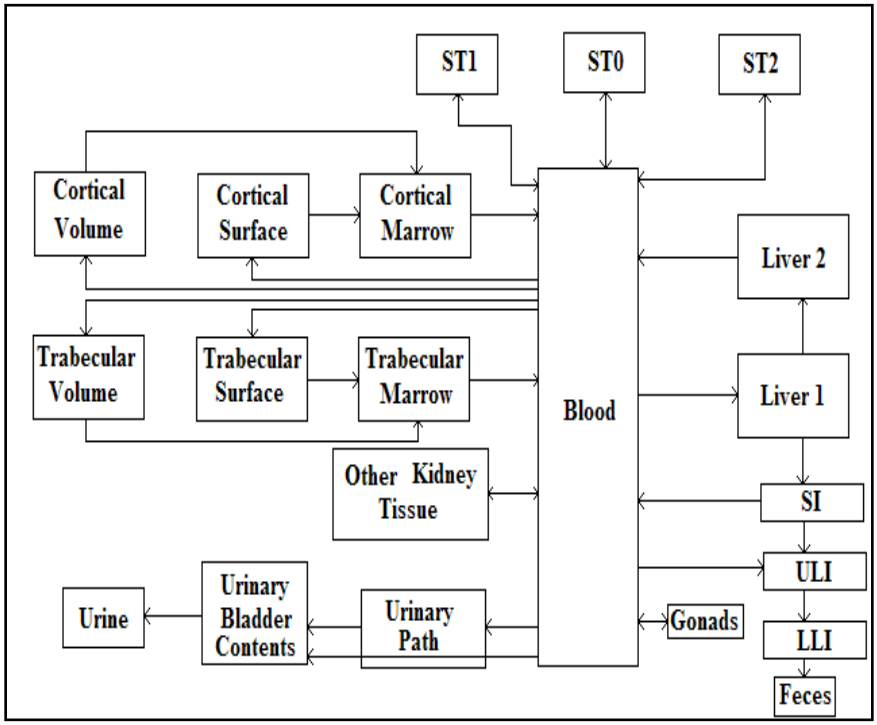


Fig. 6: Compartmentalized Luciani and Polig plutonium systemic model (Adapted from Luciani and Polig 2000)

INTERNAL DOSIMETRY CONSIDERATIONS WITH BIOKINETIC MODELS

This section summarizes the dosimetry concepts used in ICRP-30 and ICRP-60. The dosimetry models are not shown in all their detail. Specific dosimetry methods and values can be obtained from the actual ICRP-30 and ICRP-60 documents (ICRP 1979, and ICRP 1991).

ICRP-30 DOSIMETRY FOR PLUTONIUM

The ICRP-30 dosimetry method calculates the total committed dose equivalent to assess the dose to an organ or tissue over a period of 50 years after an intake of radioactive material. This is calculated using eqn (3) (ICRP 1979):

$$H_{50,T}(T \leftarrow S) = (1.6 \times 10^{-10})(U_S)[SEE(T \leftarrow S)] \tag{3}$$

where:

- $H_{50,T}(T \leftarrow S)$ = the committed dose equivalent to target organ or tissue *T* from each transformation occurring in source organ or tissue *S*
- (1.6×10^{-10}) = the conversion factor so that the committed dose equivalent can be expressed in Sv and the specific effective energy can be expressed in MeV/g
- U_s = the total number of transformations of a radionuclide occurring in the source organ or tissue *S* over a period of 50 years after intake. This value is calculated using the biokinetic models.

- $SEE(T \leftarrow S)$ = the specific effective energy imparted by the radionuclide that transformed in *S* and gets absorbed in target organ or tissue *T*

The total number of transformations occurring in the lung is calculated by the ICRP-30 respiratory tract model described earlier. The total number of transformations occurring in each organ of the GI-tract is calculated by the ICRP-30 GI-tract model. After a radionuclide passes through the respiratory or GI-tract, it is translocated to the body fluids and then to the specific tissues or organs of interest for plutonium. Clearance rates and organs of interest are given in the ICRP-30 metabolic data. The total calculated transformations for an organ is then applied to eqn (3) with the other calculated values to assess the committed dose equivalent to the target. The specific effective energy can be calculated from Eq. 4 (ICRP 1979):

$$SEE(T \leftarrow S)_j = \sum_i \frac{Y_i E_i AF(T \leftarrow S)_i Q_i}{M_T} \tag{4}$$

where:

- $SEE(T \leftarrow S)_j$ = the specific effective energy for radionuclide *j* in MeV/g
- Y_i = the yield of radiations of type *i* per transformation of radionuclide *j*
- E_i = the average energy of radiation *i* in MeV
- $AF(T \leftarrow S)_i$ = the absorbed fraction is the fraction of energy absorbed in the target organ *T* per emission of radiation *i* in source *S*. With exception to mineral bone and the GI-tract, the absorbed fractions for electrons and alpha particles

equals one when the source is the target and zero when the source is not the target (ICRP 1979).

- Q_i = the quality factor for radiation of type i
- M_T = the mass of the target organ in g

The yield and average energy of radiation used in this report for plutonium decay were taken from the nuclear decay data in the MIRD format website (Tuli and Burrows 2007). The mass of the source and target organs used for the ICRP-30 plutonium dosimetric system in this report were taken from ICRP-30. The quality factor used for alpha particles emitted by plutonium was taken from ICRP-30. The committed dose equivalent for each target organ is then used to calculate the committed effective dose equivalent using eqn (5) (ICRP 1979):

$$H_E = \sum_T w_T H_{50,T} \tag{5}$$

where:

- H_E = the committed effective dose equivalent after the total committed dose equivalent for each organ is weighted and summed
- w_T = the tissue weighting factor for the target organ T. These values are calculated by taking the ratio of the individual risk for the target T to the sum of all the risk coefficients.

The tissue weighting factors used in this report were taken from ICRP-30. The effective dose equivalent can be used to calculate the annual limit on intake (ALI) to make sure

that intakes are below the limits for occupational exposure. ICRP-30 used eqn (6) to calculate the maximum annual intake (ICRP 1979).

$$I \times H_E(per\ Bq\ intake) \leq 0.05\ \text{Sv}$$
$$I \times H_{50,T}(per\ Bq\ intake) \leq 0.5\ \text{Sv} \quad (6)$$

where:

- I (in Bq) is the annual intake of the radionuclide. The ALI is the greatest value of I which still satisfies each of the equations in eqn (6).
- 0.05 Sv = the annual limit for stochastic effects
- 0.5 Sv = the annual limit for non-stochastic effects

ICRP-60 DOSIMETRY FOR PLUTONIUM

ICRP-60 treats dosimetry very similar to ICRP-30 with few differences. The quality factor of ICRP-30 was replaced by the radiation weighting factor (w_R). ICRP-60 recommended different values than ICRP-30 for tissue weighting factors (w_T). ICRP-60 also uses the committed effective dose to assess the tissues weighted dose to the body for a period of 50 years after intake. This is shown in eqn (7) (ICRP 1991).

$$E(\tau) = \sum_T w_T \times H_T(\tau) \quad (7)$$

where:

- $E(\tau)$ = the committed effective dose

- w_T = the ICRP-60 tissue weighting factor
- $H_T(\tau)$ = the committed equivalent dose integrated over time

The committed equivalent dose is the sum of the dose to each organ over a 50 year period after intake multiplied by the radiation weighting factor. The committed equivalent dose is calculated similar to eqn (3). The number of transformations occurring in organs other than the lung and those from the ICRP-30 GI-tract were calculated using systemic models instead of the ICRP-30 plutonium dosimetry model. The ICRP-60 dosimetry method also recommended new dose limits for occupational workers. This includes an effective dose limit of 20 mSv per year averaged over five years with a further provision that the effective dose should not exceed 50 mSv in any single year (ICRP 1991). There were also non-stochastic limits placed on the lens of the eye, the skin, and the hands and feet. Radiation weighting factors and tissue weighting factors from ICRP-60 were used in this report for all combinations except that which include the full ICRP-30 plutonium dosimetry model. Even if the ICRP-30 respiratory model and GI-tract model were used in a combination that includes a systemic model other than used for plutonium in ICRP-30, the ICRP-60 dose method was still used.

UNCERTAINTY

Uncertainty in dose measurements arises from each element in the dose determination process. This includes uncertainties in measurement, assessment of intake from measurement, and assessment of dose from the intake (ICRP 1997). The uncertainty in measurement of activity, through *in vivo* measurements, excretion data, monitoring devices, etc., is the easiest stage to assess uncertainty. This is provided that

the measurements are in sufficient quantity above a decision level to reduce the error from counting statistics (ICRP 1997). Uncertainty in assessing the intake from these measurements and the dose from the intake seems to be the most difficult to quantify. It is difficult to assess the uncertainty due to an unknown time of intake and unknown individual kinetics. Reference parameters for each model are given based on probability density functions which reflect the confidence to the estimation of the parameter (ICRP 1991). ICRP-66 reference parameters have variability and uncertainty with their use. Variability is the range of values that can be used for each parameter based on the known range of each parameter. Uncertainty in each parameter refers to the range of values a parameter may have due to incomplete knowledge of the true value (Marsh et al. 2005). Since the range of the parameters can give large differences in results, the recommended values give conservative dose estimates. If enough information is available for specific individuals, then individual-specific parameters can be used to assess dose. This is especially true for transfer rates or parameters that have a large effect on dose estimates. For example, the F_1 value has a large effect on dose estimates for ingestion intakes and the absorption parameters, S_s, S_r, and f_r, along with AI transport are important for inhalation intakes. Care should be taken to collect all available information regarding these values as possible (Luciani et al. 2001). For cases with large amounts of data in which the time of intake is known there is a step-by-step procedure to give guidance on the order of which parameter values should be varied when analyzing measurements (Marsh et al. 2005).

For the reasons mentioned above, an estimate of the level of uncertainty when assessing an intake is difficult to achieve and the reference parameters used should give a nominal value of intake (ICRP 1997). Then each contributing factor to the uncertainty may be analyzed according to the situation.

CHAPTER III

MATERIALS AND METHODS

SIMULINK COMPUTER MODELING

Simulink is a software package used to model dynamic systems. Simulink provides a graphical user interface that allows differential equations to be modeled as block diagrams that are connected as one would draw on paper. This is a useful tool for modeling the transfer of radionuclides throughout the body as a system of coupled differential equations. This code uses subsystems so that the user can view the entire model from the top level or double click on a subsystem to see increasingly more detail in lower levels. Computer modeling is necessary for this research as each system (respiratory, GI, and systemic) is represented by large numbers of coupled, first-order differential equations. The Simulink program used in this research was included with MATLAB and Simulink student version package 7.4.0 R2007a.[1] Among the advantages of using Simulink to model radionuclide transfer throughout the body compared to commercial dose calculating packages are the capability of altering the intakes to allow for fluctuating or chronic intakes, and the ability to change transfer parameters for specific individual kinetics.

Figs. 7-9 show a sample model used for this research. This is included to show the subsystem and block diagram method that Simulink used to solve the coupled, differential equations describing radionuclide transfer. Figs. 7-9 describe the ICRP-30 GI-tract system. Fig. 7 shows the complete ICRP-30 GI-tract system from the top level.

[1] MATLAB & Simulink Student Version: Getting started with Simulink user manual

This includes the inputs for radionuclide inflow from the respiratory tract, the F_1 value specific for the radionuclide being modeled, the initial activity entering the GI-tract, and the radionuclide half life. This diagram also shows output displays for the activity and the number of transformations occurring in each compartment of the GI-tract. Each of the subsystems of the GI-tract (stomach, small intestine, upper large intestine, and lower large intestine) is shown in Fig. 8. This diagram is seen by double clicking on the main, GI-track system block shown in Fig. 7. Fig. 8 shows how the output of one subsystem is connected to the input of another subsystem to model the flow of radionuclides from one compartment to another. This couples the differential equations that describe each subsystem. Fig. 9 shows the lowest block diagram level used to solve the differential equation of the stomach subsystem. The block diagram in Fig. 9 is obtained by double clicking on the stomach subsystem in the block diagram of Fig. 8. The block diagram, shown in Fig. 9, is used to solve eqn (8):

$$\frac{dq_i(t)}{dt} = \dot{I}(t) + \sum_{\substack{j=1 \\ j \neq i}}^{n} \lambda_{j,i} q_j - (\lambda_R + \sum_{\substack{j=1 \\ j \neq 1}}^{n} \lambda_{i,j}) q_i \tag{8}$$

where: $q_{st}(t)$ is the activity in the stomach at time t, λ_{st} is the biological clearance rate from the stomach compartment to the small intestine compartment, λ_r is the radioactive decay constant of the radionuclide, and $\dot{I}$ is the rate of radionuclide transfer from the respiratory tract to the GI-tract.

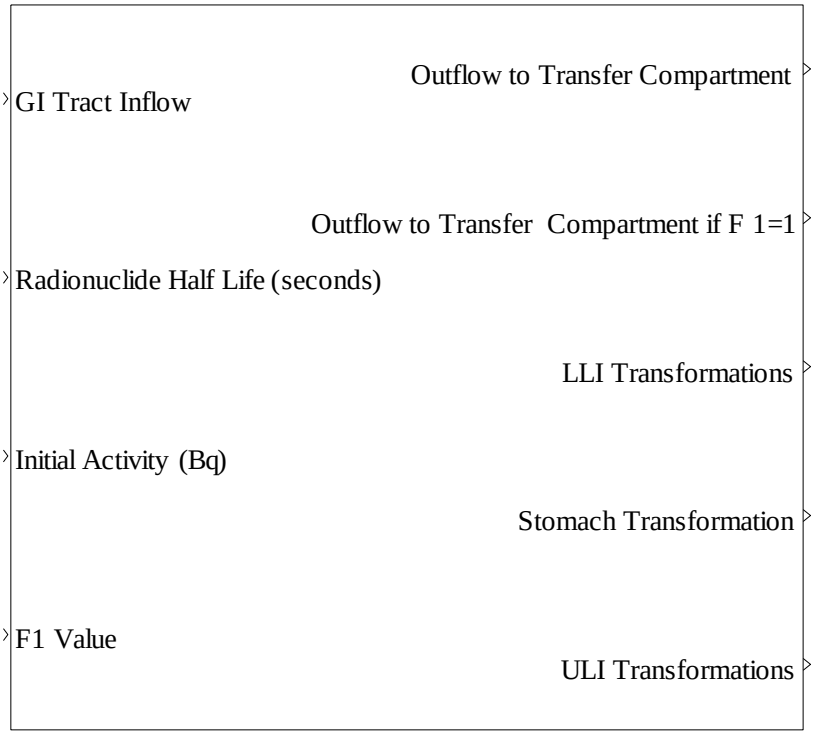


Fig. 7: Top level block diagram of the ICRP 30 GI-tract model

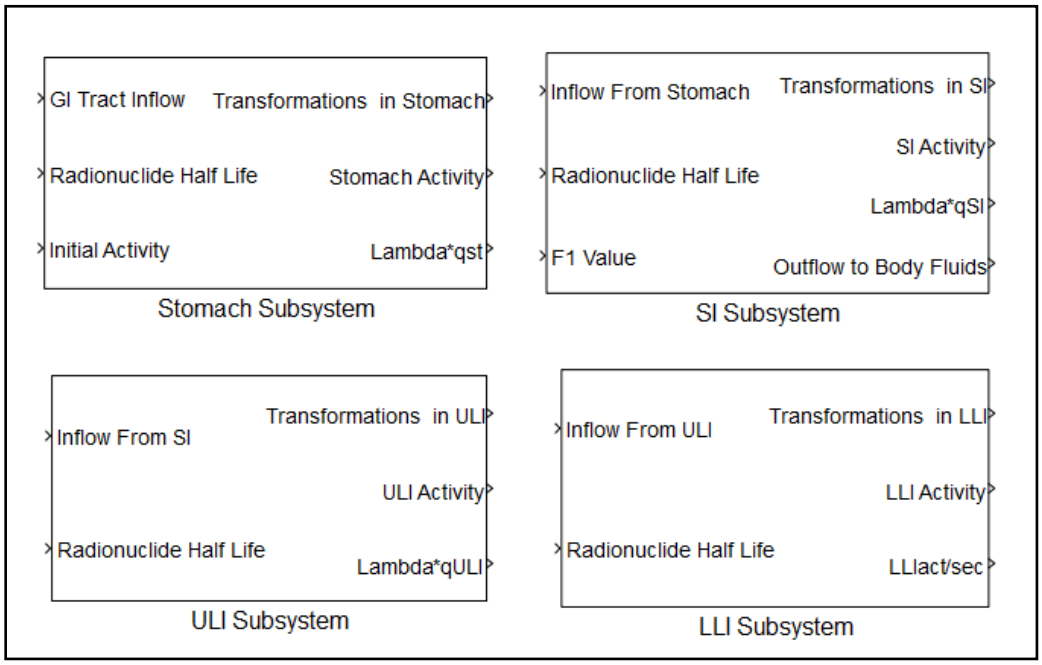


Fig. 8: Subsystem block diagram showing each compartment of the GI-tract model

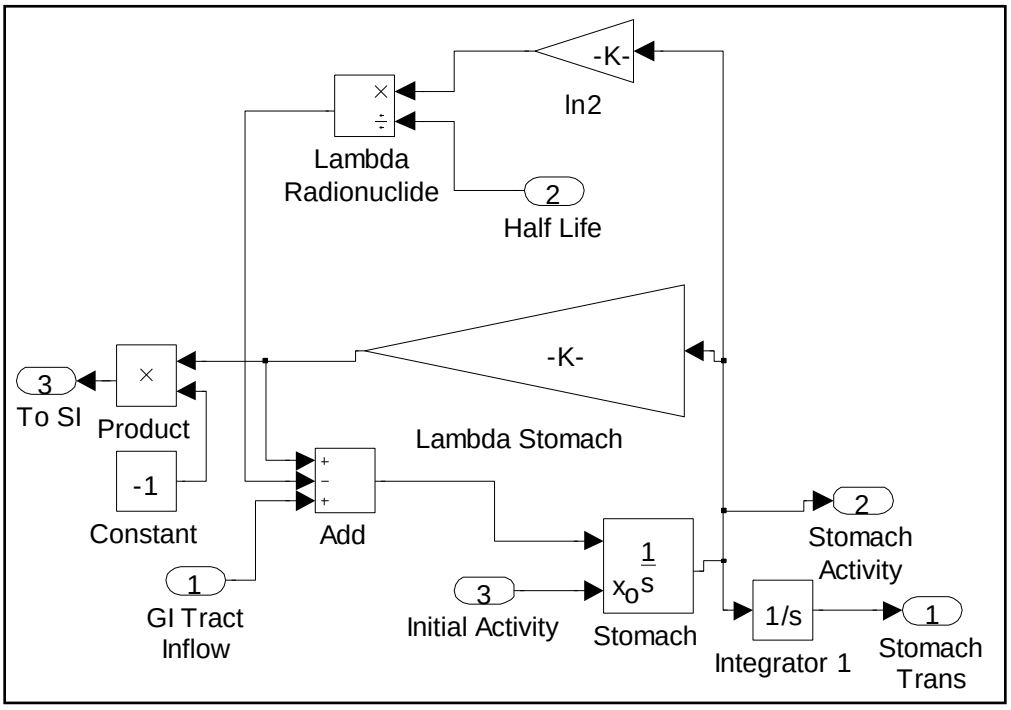


Fig. 9: Lowest level block diagram of the stomach subsystem

PROCEDURES

To meet the objectives, the ICRP-30 GI-tract model, the ICRP-30 respiratory tract model, the ICRP-66 respiratory tract model, the ICRP-67 plutonium systemic model, and the Luciani and Polig plutonium systemic model were put into block diagrams in similar fashion to Figs. 7-9 using Simulink. These models were used for excretion analysis and dosimetry purposes. The ICRP-30 plutonium dosimetry model was also built for dosimetry comparisons with each combination that used ICRP-60 dosimetry values. The above biokinetic models were used to calculate total radionuclide transformations in tissues or organs of interest. Specific configuration parameter settings were used with each model and are important to keep constant for each model to

allow reproducibility. A variable-step type solver was used so that the step-size changes from step to step, allowing smaller steps when the model state changes rapidly and larger steps when the model state changes slowly. This reduces the simulation time; especially when the activity transfer rate levels off over time. Each model used the ode45 (Dormand-Prince) solver. This is the default solver for variable-step models in Simulink. If any of the simulations failed to run, a different solver could be chosen, but this did not occur in any of the simulations for this research. The relative and absolute tolerance levels were chosen to be low (10^{-16}) because using the default tolerances gave inaccurate results at low values due to a higher acceptable error. Decreasing the tolerance increases the simulation time, but improved the results significantly.

After each system was modeled in Simulink, the systems were combined for statistical comparisons. The combinations used to represent the human body in this study and compare excretion results were:

1. Combined ICRP-30 respiratory tract model, ICRP-30 GI-tract model, and ICRP-67 plutonium systemic model. (Combination 1)
2. Combined ICRP-66 respiratory tract model, ICRP-30 GI-tract model, and ICRP-67 plutonium systemic model. (Combination 2)
3. Combined ICRP-30 respiratory tract model, ICRP-30 GI-tract model, and Luciani and Polig plutonium systemic model. (Combination 3)
4. Combined ICRP-66 respiratory tract model, ICRP-30 GI-tract model, and Luciani and Polig plutonium systemic model. (Combination 4)

Once these models were built, the ^{239}Pu specific properties were added to each (ICRP 1997; Tuli and Burrows 2007). These included a half-life of 24,110 years, an F_1 value of 10^{-5}, an initial activity of zero Bq, and the biological decay constants used for each compartment. The default form of plutonium (oxide form as PuO_2) was used in this study. Since the size of particles were unknown, 1 μm AMAD sized particles were used with combinations which included the ICRP-30 respiratory tract model and 5 μm AMAD sized particles were used with combinations which included the ICRP-66 respiratory tract model. Each model was set up so that the daily urinary and fecal excretion could be plotted versus time. Each of the above models' excretion and dose values were checked and matched well with published results (ICRP 1979, ICRP 1994, ICRP 1997, Bair 1995, Potter 2002).

After the models were built, the daily urinary and fecal excretion amounts were plotted versus time, and a comparison was made for each mode of intake (inhalation, ingestion, and wound). This was done to assess the possibility of interchanging models when assessing intakes. Each model used a 1 Bq intake of ^{239}Pu for comparison.

After the comparison, the models were applied to the case study to assess the workers intake. Not enough biokinetic information or excretion data are available for the worker so using individual-specific parameters are not an option to study this intake. Reference parameters given for each model were used throughout the investigation. The mode and time of intake was established based upon the excretion and area monitoring data. This was the most difficult and error prone task, as the positive bioassay results were unexpected, and neither the mode nor the time of intake was known. To assess the

mode of intake, the fecal excretion measurements from the worker along with the error in each measurement were modeled versus time, with the first data point set as day one. This was done to compare the worker's fecal excretion measurements to the excretion output for the biokinetic models in Simulink, and to get a general idea of which mode of intake gave the correct form of daily excretion rates. Combination 1 was used to judge the assumed mode of radionuclide intake because the ICRP-30 respiratory and GI-tract models are currently the recommended models by the U.S. NRC (U.S. NRC 1993). These models were combined with the ICRP-67 plutonium systemic model so that the excretion data could be modeled. Even though Combination 1 was used to compare to the workers excretion data, it should not matter which combination was used because they each show the same general form in excretion results and an exact match is not needed for comparison to the workers data. The model was simulated with different modes of intake (ingestion, wound, and inhalation) to get an idea of which could fit the workers bioassay data best. Once the mode of intake was assumed, the time of intake had to be decided upon. This was done based on the time the worker was at the plant and the area monitoring data.

Once the mode and time of intake were decided upon, each of the combinations were used to compare the total intake of plutonium. Eighteen simulations were run for each combination (three for each of the worker's six positive fecal measurements) so that the model excretion results match each of the worker's excretion measurements and upper and lower measurement error values. For example, Combination 1 was used with the assumed mode and time of intake. One simulation was run with a certain intake, so

that the excretion results from the model would match the upper limit of the error bar for the first bioassay data point of the worker. Another simulation was run with an intake amount so that model excretion results would match the lower limit of the error bar for the first bioassay data point of the worker. A third simulation was run with an intake amount so that model excretion results would match the first bioassay data point of the worker. These same steps were implemented for the next data point for the worker, and so on. These steps allowed the intake to be calculated using a minimized chi-squared fit, weighted by the inverse of the variance for each excretion data point. The equations used are shown in eqn (9). This method was implemented with each of the other combinations to compare the intakes that would be calculated (Knoll 2000).

$$\langle x \rangle = \frac{\sum_{i=1}^{N} a_i x_i}{\sum_{i=1}^{N} a_i} \tag{9}$$

$$a_j = \frac{1}{\sigma_{x_j}^2}\left(\sum_{i=1}^{N} \frac{1}{\sigma_{x_i}^2}\right)^{-1}$$

$$\frac{1}{\sigma_{\langle x \rangle}^2} = \sum_{i=1}^{N} \frac{1}{\sigma_{x_i}^2}$$

where: $\langle x \rangle$ is the best value of the assessed intake, x_i is an individual intake to match a single worker excretion point, a_i is the weighting factor to be given to each individual excretion point, N is the number of excretion data points, σ_{x_i} is the error in the intake that matches a single excretion points error limit, and $\sigma_{\langle x \rangle}$ is the error in the best assessed intake value based on propagating the error in each excretion measurement. This

assesses the best intake by giving more weight to the excretion measurements with smaller error.

CHAPTER IV

RESULTS

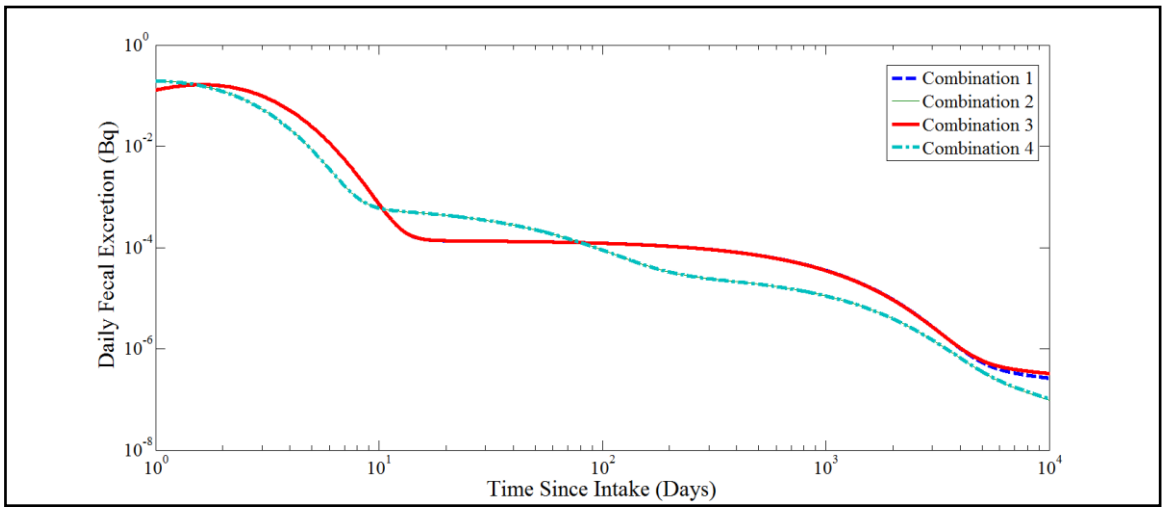
COMPARISON OF EXCRETION RATES

Each of combinations 1-4 were simulated for acute inhalation, ingestion, and wound modes of intakes of 1 Bq of ^{239}Pu to compare the predicted daily fecal and urinary excretion values. Figs. 10-12 show how the predicted daily fecal and urinary excretion rates change with time. Fig. 10 shows the daily excretion rates for an acute inhalation mode of intake. Fig. 10A shows that Combinations 1 and 3 gave similar fecal excretion rates and Combinations 2 and 4 gave similar fecal excretion rates. This is because the respiratory models use different retention times and transfer rates to the blood or GI-tract. The fact that Combinations 1 and 3 and Combinations 2 and 4 gave similar fecal excretion rates is because the GI-tract is responsible for the majority of the fecal excretion up to about 4,000 days for plutonium. This is due to the small F_1 value for plutonium, which indicates a small fraction of the radionuclides passing through the GI-tract goes to the blood. Combinations 2 and 4 have no noticeable differences in the daily fecal excretion rates for 10,000 days post intake. Fig. 10B shows that each combination gives a different urinary excretion rate as a function of time. The daily urinary excretion rate in Fig. 10B is orders of magnitude lower than the daily fecal excretion rate for each combination. This is why bioassay by fecal analysis is preferred over urinary analysis for this class of inhaled radionuclide. Radionuclides must pass through the systemic model before being excreted through urine, while most of the excreted activity through feces travels from the respiratory model directly to the GI tract

model. Fig. 10B also shows that the ICRP-30 respiratory tract model transfers more activity to the systemic model than the ICRP-66 respiratory tract model by comparing Combination 1 to Combination 2 or by comparing Combination 3 to Combination 4. Using a combination with the ICRP-30 respiratory system will give a higher urinary excretion rate than using a combination with the ICRP-66 respiratory system. This figure also shows that using a combination with the ICRP-67 plutonium systemic model will give a higher urinary excretion rate than using a combination with the Luciani and Polig plutonium systemic model. This could be in part to the transfer of activity from the ST1 compartment to the urinary bladder contents compartment of the ICRP-67 model.

Fig. 11 shows the daily excretion rates for an acute ingestion intake of plutonium. Combinations 1 and 2 and Combinations 3 and 4 give the same excretion rates for 10,000 days after intake. This is because the respiratory models are not important for a pure ingestion intake and the choice of respiratory model is irrelevant. The daily fecal excretion rate, shown in Fig. 11A, is very similar for each combination and implies that the fraction of fecal excretion coming from the ICRP-67 and Luciani and Polig systemic models are in close agreement for ingestion intakes. The similarity is also because most of the fecal excretion comes from material passing through the GI-tract for plutonium and the fecal excretion from material passing through the systemic system has a lesser role. Fig. 11B shows that the daily urinary excretion values are higher when a combination uses the ICRP-67 plutonium systemic model as opposed to the Luciani and Polig plutonium systemic model (same as with inhalation intake).

A:



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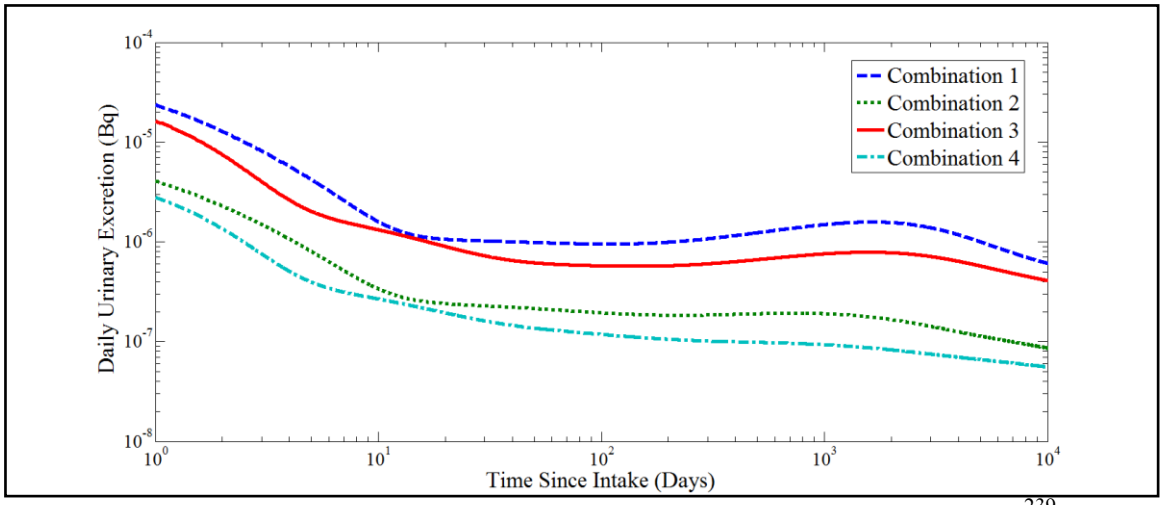
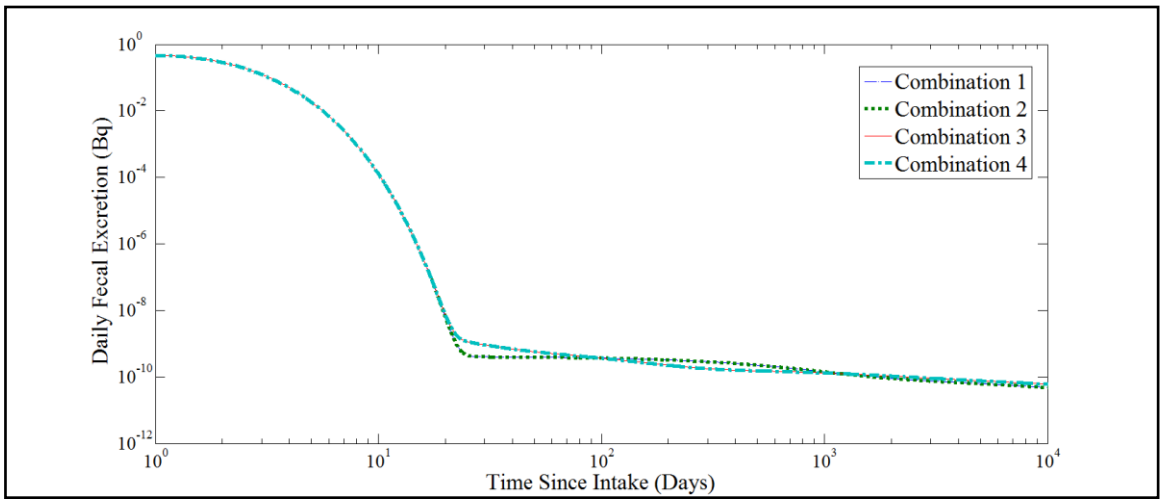


Fig. 10: Daily excretion rates versus time for acute inhalation of one Bq of ^{239}Pu.
A: Daily fecal excretion rate B: Daily urinary excretion rate

A:



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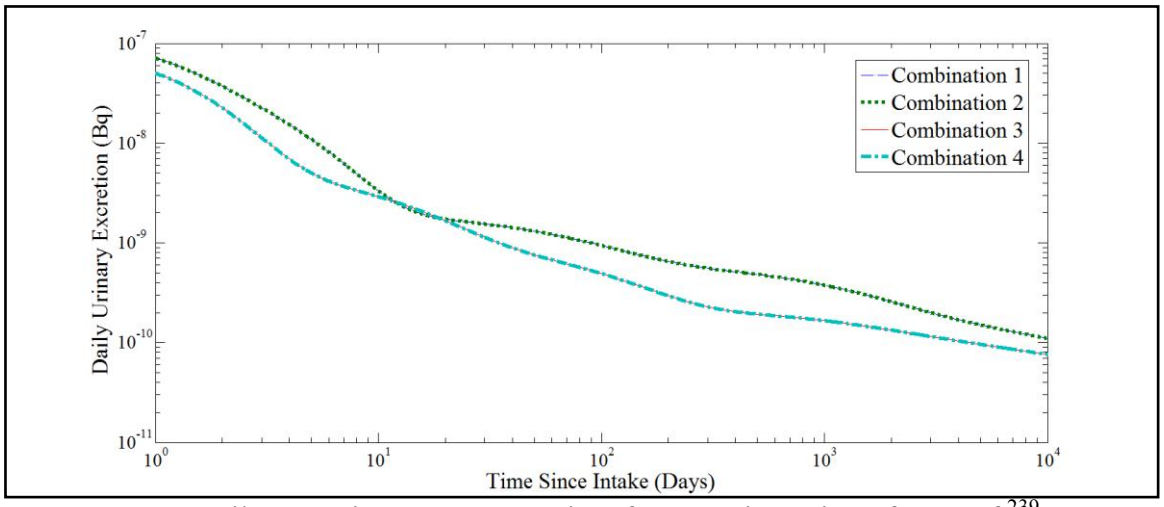
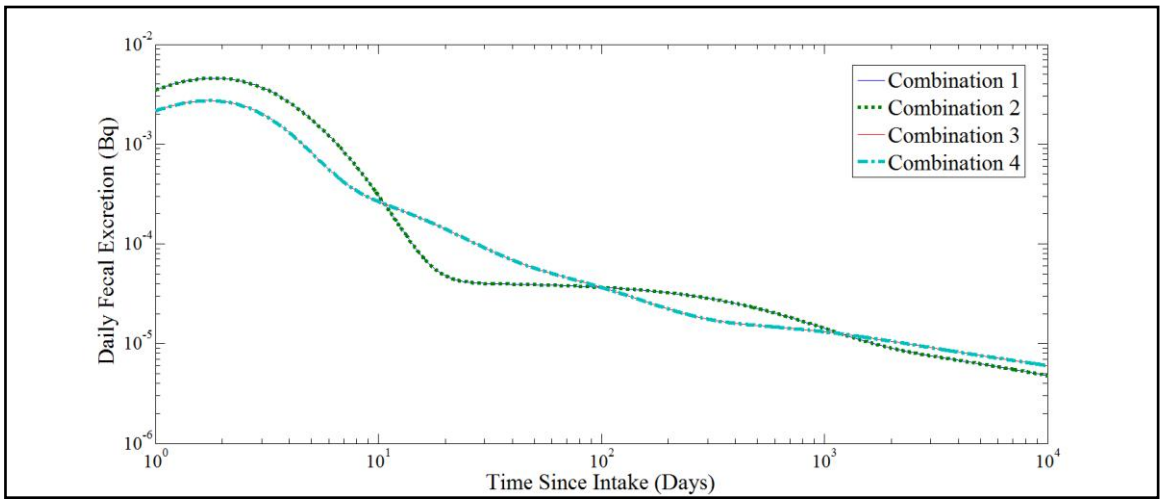


Fig. 11: Daily excretion rates versus time for acute ingestion of 1 Bq of ^{239}Pu
A: Daily fecal excretion rate B: Daily urinary excretion rate

A:



B:

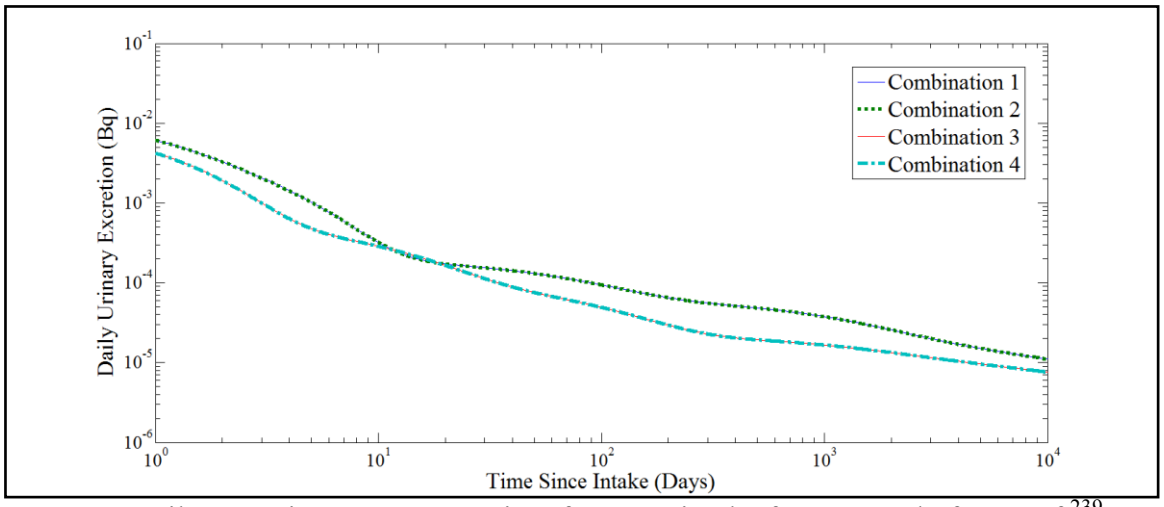


Fig. 12: Daily excretion rates versus time for acute intake from wound of 1 Bq of ^{239}Pu
A: Daily fecal excretion rate B: Daily urinary excretion rate

Fig. 12 shows the daily excretion rates for an acute intake from a wound or injection. Figs. 12A and 12B show that combinations 1 and 2 have no noticeable differences in the daily excretion rates and combinations 3 and 4 have no noticeable differences in the daily excretion rates for 10,000 days post intake. This is as expected because only the GI-tract and systemic models are important for this mode of intake.

Fig. 12B shows that again the daily urinary excretion values are higher when the combination uses the ICRP-67 plutonium systemic model as opposed to the Luciani and Polig plutonium systemic model. Fig. 12 also shows that unlike the inhalation and ingestion modes of intake, the excretion rates for feces and urine are of similar orders of magnitude when there is an intake from a wound. This is important because it shows that bioassay measurements using urine would be important for this mode of intake.

CASE STUDY RESULTS

Combination 1 was used to decide upon the correct mode of intake for the worker. From the workers fecal bioassay data, it was obvious that an ingestion mode of intake could be ruled out because the predicted daily fecal excretion rate drops almost ten orders of magnitude after ten days, which did not match the worker's fecal bioassay measurements. Wound and inhalation intakes have fecal excretion rates that drop two and three orders of magnitude, respectively, over a period of about 10 days, but the worker's daily fecal excretion data drops less than an order of magnitude in 10 days. A chronic or fluctuating intake could explain the lower slope in the worker's excretion data, however, it is unlikely to have a chronic intake from a wound over a long period of time. Also, Fig. 12 shows that an intake from a wound would show positive urine bioassay results that would give similar excretion rates to that of the fecal bioassay results. There was only one positive urine sample of the worker for ^{239}Pu and this value was approximately two orders of magnitude lower than the fecal bioassay results, so it can be assumed that the worker did not solely have an intake from a wound. This leads to the assumption of a chronic inhalation mode of intake over a certain time period. This

is consistent with air monitoring data because the daily activity detected was relatively unchanging when the worker was in areas containing radioactivity. There was a spike in the worker's bioassay data about 60 days after the first measurement. This occurred when the worker was placed on restriction outside of radioactive work areas. This can only be explained by a separate intake from the first assumed chronic inhalation intake. This is consistent with either an acute inhalation intake, a short chronic inhalation intake, or an ingestion intake. Area monitoring data did show elevated daily activity detected in areas where the restricted worker could have been at this time (areas with no radiation work). Other workers also showed similar levels in their bioassay measurements during this time frame. Based on the elevated levels read by the area monitors over a period of about five days, a five day chronic inhalation intake is assumed during this time period.

The time period of the intake is much harder to estimate if it is unknown, especially for chronic intakes. This is because daily excretion rates from chronic intakes tend to level off until the chronic intake ceases. Many more bioassay data points would be needed to get a better idea of the time of intake. Also, many urine bioassay data points could also be beneficial so that the urinary daily excretion could be modeled, but the measurements must be very sensitive for this class of radionuclide. The time of intake can be judged based on the worker's time of employment at the facility and the area monitoring data. Although the area monitors should not be used to assess a quantitative intake amount because of the dependence on monitor location, they should be used to get a general trend as to the fluctuations in the amount of measured radiation over time. This can give a good indication to the time frame of intake. The facility had

relatively constant monitoring data over time when the worker was in areas containing radioactivity, which is consistent with a chronic mode of intake. It has recently been shown that in situations where an intake has occurred in a monitoring interval and there is no indication about the actual time of intake, then the only unbiased estimate of intake is obtained by assuming a constant chronic intake throughout the monitoring period (Birchall et al., 2007). This leads to the assumption that there was a constant chronic intake since the worker started working in the monitored waste area. It is also assumed that the worker did not receive any intake when placed outside of radiological areas for a period of two months in the middle of the work period.

As stated above, it was assumed that the worker has two separate chronic inhalation intakes, one since the worker started in areas containing radioactive materials and ending when the worker was placed on work restriction, and the other beginning two months after the worker was placed on restriction and lasting for a period of five days. This fluctuating intake was input using 'if' and 'else' blocks. Each model combination was used to assess the intakes to the worker based on the fecal excretion measurements. Since there were two separate chronic intakes, the chi-squared method was used with the first three bioassay measurements to assess the first chronic intake. Once the original chronic intake was assessed, this value was used in the intake input and the chi-squared method could be applied to the final three bioassay measurements to assess the second chronic intake. Fig. 13 shows the calculated intakes based on the chi-squared method for each combination. This shows the calculated initial chronic intake and the second calculated chronic intake when the worker was placed on restriction. Fig. 13 shows that

Combinations 1 and 3 give similar intakes as well as Combinations 2 and 4. This is due to the fact that Combinations 1 and 3 use the ICRP-30 respiratory tract model and Combinations 2 and 4 use the ICRP-66 respiratory tract model. Combinations using the ICRP-30 respiratory tract model gave a smaller initial chronic inhalation intake, but a larger final inhalation chronic intake compared to combinations using the ICRP-66 respiratory tract model. The uncertainty shown in Fig. 13 is not the overall uncertainty in the intake, but the uncertainty due to the propagation of the error in the measured activity from the fecal bioassay data. It is tough to quantify the total uncertainty of the intake in this case where the time and complete mode of intake are unknown. Fig. 14 shows the calculated target functions for each assumed intake. The target function is a measure of the deviation of the measured excretion values from the theoretically predicted values (Luciani and Polig 2000). If more excretion data points were given or if the kinetic information of the worker were better understood then the target function could be used to optimize model parameters. Fig. 14 shows that the initial assumed chronic inhalation intake gives a much higher deviation than the second assumed chronic inhalation intake. Fig. 14 also shows that combinations using either of the systemic models gave the same deviation as long as the respiratory tract model was held constant. This is not the case for the respiratory model as it seems that combinations involving the ICRP-66 respiratory tract model deviate much more with the assumed initial chronic inhalation intake. However, combinations using the ICRP-66 respiratory tract model deviate less for the final assumed chronic intake.

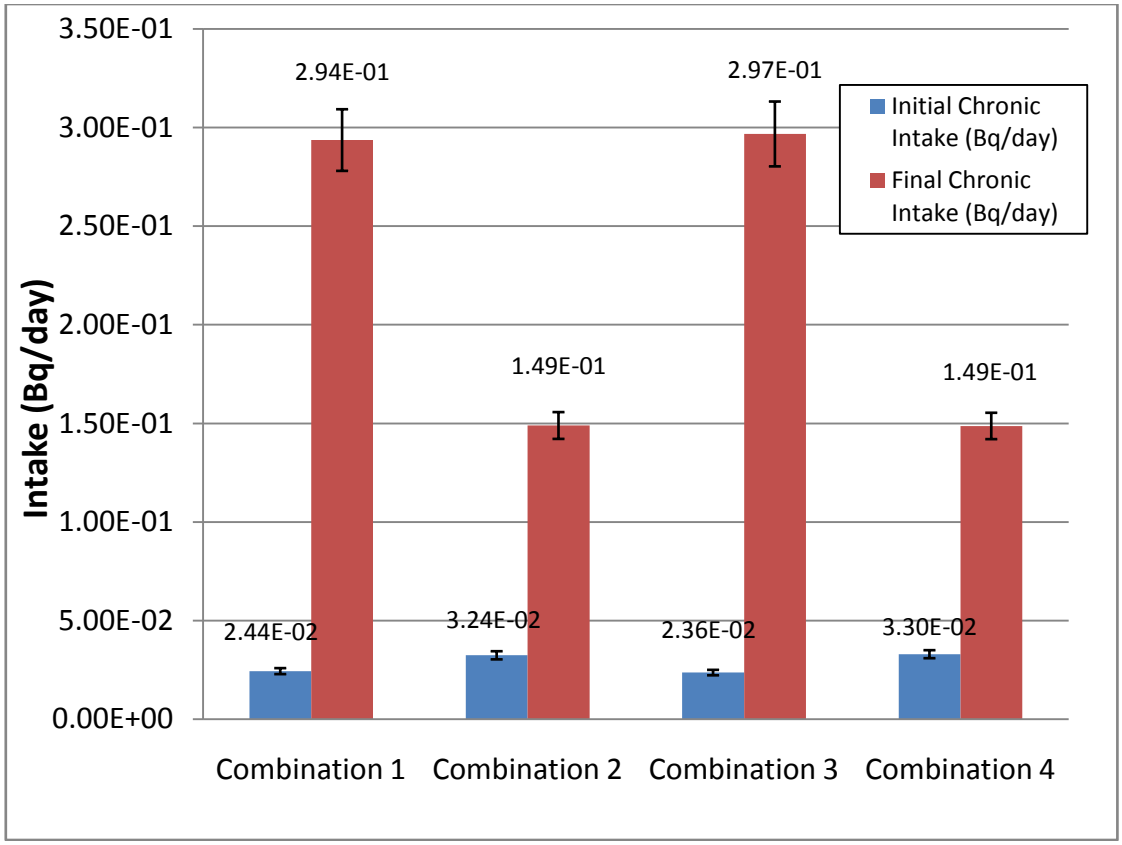


Fig. 13: Calculated intake rates (Bq/s) for each combination

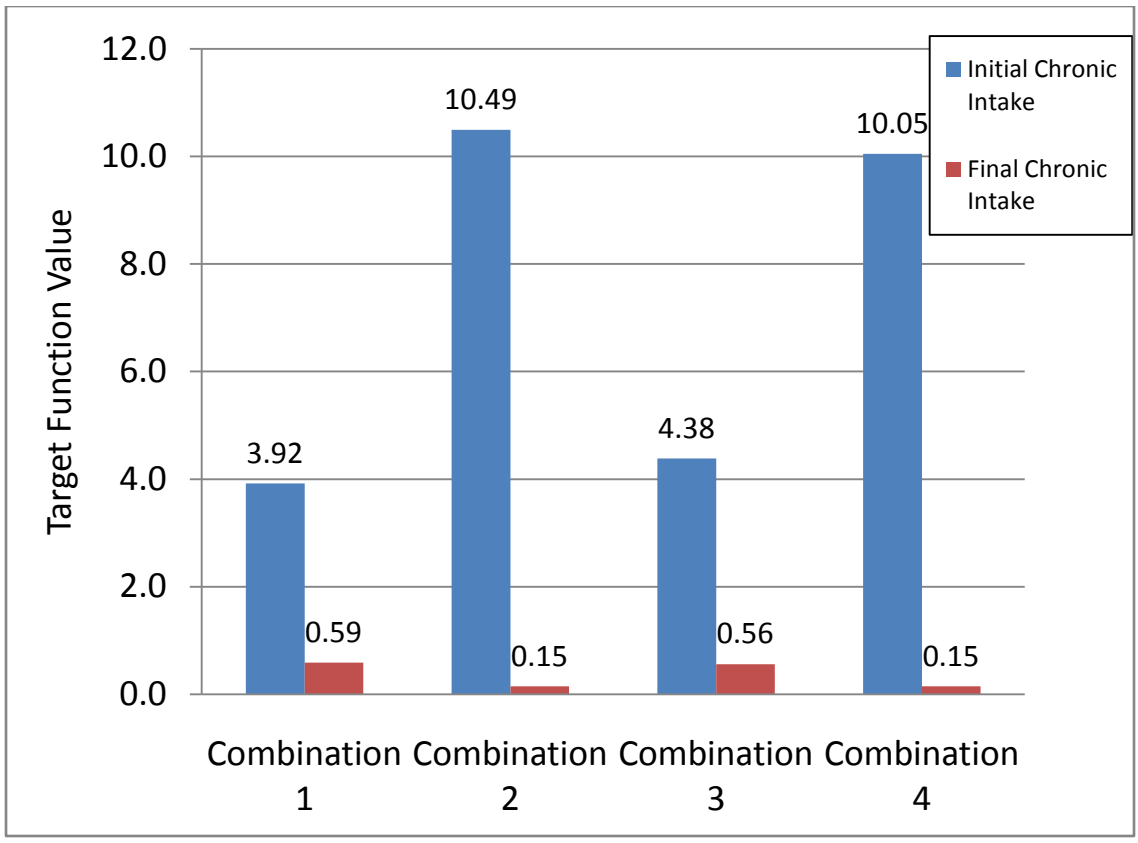


Fig. 14: Calculated target functions for each intake

The commited effective doses from ICRP-60 and committed effective dose equivalents from ICRP-30 for the calculated intakes using the combinations are given in Table 1. The combinations using the ICRP-30 respiratory tract model give a much higher commited effective dose as opposed to those using the ICRP-66 respiratory tract model. This is due to the fact that the ICRP-30 respiratory tract model gives about an order of magnitude higher dose per Bq intake than the ICRP-66 respiratory tract model. However, the combinations with the ICRP-66 respiratory tract model calculate a higher total radionuclide intake. This is noticeable when the ICRP-30 plutonium dose model was used to calculate committed effective dose equivalents for the assessed intakes each

of the combinations provided. Table 1 shows the calculated committed effective dose equivalent values using the ICRP-30 plutonium dosimetry model for each of the assessed intakes. The ICRP-30 dosimetry is widely used today due to its simplicity and Table 1 shows its comparison with the calculated committed effective doses using the assessed intakes from the different combinations.

Table 1: Doses calculated by each combination for the intake assessed

	Committed Effective Dose (Sv)	**Committed Effective Dose Equivalent (Sv)**
Combination 1	0.00098	0.00140
Combination 2	0.00018	0.00175
Combination 3	0.00103	0.00135
Combination 4	0.00019	0.00177

CHAPTER V

CONCLUSIONS

MODEL COMPARISONS

Based on Fig. 10A, it is apparent that any combination will give a similar form of the daily fecal excretion for an acute inhalation intake of plutonium. However, interchanging respiratory models to assess an intake can lead to large differences in predicted excretion. The predicted fecal excretion by interchanging respiratory models for an inhalation intake has up to a 110% difference for the first 100 days post intake and up to 185% difference after 100 days. Due to the complexity of the ICRP-66 respiratory tract model, it would be much simpler if the ICRP-30 respiratory tract model could be used to assess an intake from fecal excretion data. Fig. 10A shows that using the ICRP-30 respiratory tract model can be sufficient to assess an intake depending on the time post intake. This respiratory model can be used with either the ICRP-67 or the Luciani and Polig plutonium systemic models when assessing an intake from fecal bioassay data as there is little change in excretion rates when interchanging systemic models. The systemic models are not as interchangeable when assessing an intake based on urine bioassay data, as shown in Fig. 10B. The predicted urine excretion rate can be over an order of magnitude different from another combination when interchanging the respiratory models. The predicted urine bioassay results can have up to 75% difference by interchanging the systemic models. However, fecal bioassay is best to assess an intake for this class of radionuclide.

Fig. 11A shows the predicted fecal bioassay results for each combination when there is an ingestion intake. Choice of respiratory model is not important for an ingestion intake. The largest difference in predicted fecal excretion results occurs during a period from 20-40 days post intake (up to 90% difference). However, the systemic models can be interchangeable at any other times post intake when using fecal analyses. Again, care should be taken into consideration when assessing an intake using urine bioassay measurements. Using the Luciani and Polig plutonium systemic model will give lower urine excretion rates compared to the ICRP-67 plutonium systemic model at almost any time post intake (up to 90% difference). This is noticeable for the ingestion intake shown in Fig. 11B.

Fig 12 shows predicted daily excretion results from wound or injection intakes. Again, the interchangeability between respiratory models is unimportant for this mode of intake. There is up to 50% difference between fecal results within the first 10 days post intake and up to 95% difference after 10 days. It is important to take this into consideration when deciding which systemic model to use at specific times post intake. For fecal bioassay measurements, a combination using the Luciani and Polig plutonium systemic model should be used because it matches better with actual studies during this time frame after an intake from a wound (Luciani and Polig 2000). The predicted daily urine excretion from the combinations is similar in form to that of an ingestion intake. A combination should be used with the Luciani and Polig systemic model when assessing an intake from urine bioassay data because the ICRP-67 plutonium systemic model tends to overestimate the predicted urine excretion rate (Luciani and Polig 2000).

CASE STUDY

The case study used for this research shows the difficulty in assessing an intake when the mode, amount, or time or intake is unknown. Once an assumed mode and time frame of intake were decided upon, each combination was used to assess the intake. It was concluded that the worker had two separate chronic inhalation intakes. This conclusion was based upon the fecal bioassay measurements and the air monitoring data. The results of the calculated intakes are shown in Fig. 13. These results show that for these chronic inhalation intakes, the estimated intake will differ by about 3% when interchanging the two plutonium systemic models. However, the predicted intakes differ by about 30% when interchanging the ICRP-30 or the ICRP-66 respiratory tract models. This is due mainly to the differences in the rate of radionuclide transfer from the respiratory tract model to the GI-tract model. The calculated target functions for each intake are shown in Fig. 14. It was shown that the assumed initial chronic inhalation intake deviated the most from the measured excretion values for each combination compared to the second chronic inhalation intake. There are many reasons that could explain this deviation:

- The mode of intake may not be entirely from a chronic inhalation intake. There might also be a small intake from the laceration the worker received that would affect the excretion measurements. It would be hard to assess an intake when there are competing modes.

- Some of the worker's excretion measurements may be outliers. With so few excretion measurements, an outlier would greatly affect the calculated target function and the assessed intake amount.
- The exact kinetics of the individual worker may not match well with the standard parameters given by the biokinetic models used in the combinations. This would affect the rate of excretion of activity from the body.
- The assessed initial chronic inhalation intake may be wrong. However, given the small amount of bioassay data, the assessed intake seems most probable.

The second assumed chronic inhalation intake, after the worker was placed on restriction, seems to fit each combination very well. This can be seen in the calculated target functions of Fig. 14. It is unclear why the assessed second chronic intake fits the worker's excretion measurements much better. It could be due to the fact that the time period of the second intake was much smaller than the first (a few days as opposed to a few years). The reference parameters used in each biokinetic model were mostly based upon acute intakes from studies as opposed to chronic intakes.

The results of the calculated doses from the assessed intakes are shown in Table 1. The table shows that interchanging the systemic models for this assumed chronic intake gave a difference of dose of up to 10%. The differences in doses calculated by interchanging the respiratory models are almost an order of magnitude different. The differences in calculated doses are both due to the different intake assessments and differences in how each model is used to calculate dose. The lung dose is calculated differently using the ICRP-30 respiratory model and the ICRP-66 respiratory model.

Table 1 also shows the calculated committed effective dose equivalent for each assessed intake using the plutonium ICRP-30 dosimetric model. The largest difference in calculated dose from the assessed intakes is about 30% using this dosimetry model. It may be much more efficient to assess an intake from bioassay data using one of the model combinations in this study followed by the use of the ICRP-30 dosimetric model to assess the committed effective dose equivalent. The ICRP-30 dosimetric model cannot be used to assess an intake based on excretion data (since it does not give specific excretion pathways), but is a computationally efficient model to assess dose once the intake is known.

FUTURE WORK

There are many possible ways that the methods used in this research could be expanded or improved. Some examples of possible future work include the following:

- More case studies - This research covered the comparison and interchangeability of models used to assess intakes and dose. The models were then used to assess an unknown intake for a case study. It would be beneficial to use the same methods for other case studies involving different modes of intakes with more bioassay measurements. Studying the interchangeability of these models for only one case study cannot provide any conclusive results for other intake modes. The interchangeability of different models with ingestion or injection intake modes is also of importance. It would also be meaningful to test these models on known intakes (including mode, time frame, and amount of intake) to legitimize the results.

- Inclusion of more biokinetic models - This research could also be expanded to include more biokinetic models. This can include alternative GI-tract models, respiratory tract models, or systemic models. Also, other radionuclides could be included in the study. The ability for certain models to use alternative parameters, like the ICRP-66 respiratory tract model, specific to an individual for whom monitoring data are available could be addressed if enough bioassay measurements are taken and the exact intake is known.
- More research involving chronic inhalation intakes - The results presented in this research regarding the strong deviation from model predictions for long chronic inhalation intakes (as shown by the target function) could be studied more in detail. This type of study would need to include known chronic inhalation case studies and acute inhalation case studies. Comparisons could be made about the deviations from model predictions. It may turn out that these biokinetic models do not fit well with chronic inhalation intakes over long periods of time.

REFERENCES

Bair WJ. The ICRP human respiratory tract model for radiological protection. Radiat Protect Dosim 60(4):307-310; 1995.

Birchall A, Puncher M, Marsh JW. Avoiding biased estimates of dose when nothing is known about the time of intake. Radiat Protect Dosim. In press.

Eve IS. A review of the physiology of the gastrointestinal tract in relation to radiation doses from radioactive materials. Health Phys 12: 131-161; 1966.

Cember H. Introduction to health physics. 3rd ed. New York: McGraw-Hill; 1996: 544-545.

Dolphin GW, Eve IS. Dosimetry of the gastrointestinal tract. Health Phys 12: 163-172; 1966.

International Commission on Radiological Protection (ICRP). Report on committee II on permissible dose for internal radiation. New York: Pergamon; ICRP Publication 2; Adopted July 1959.

International Commission on Radiological Protection (ICRP). Limits for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publication 30 Part 1; Ann ICRP 2(3/4); 1979.

International Commission on Radiological Protection (ICRP). Age dependant doses to members of the public from intake of radionuclides: Part 1. Oxford: Pergamon Press; ICRP Publication 56; Ann ICRP 20(2); 1989.

International Commission on Radiological Protection (ICRP). Recommendations of the International Commission on Radiological Protection. Oxford: Pergamon Press; ICRP Publication 60; Ann ICRP 21(1-3); 1991.

International Commission on Radiological Protection (ICRP). Age dependent doses to members of the public from intake of radionuclides. Oxford: Pergamon Press; ICRP Publication 67; Ann ICRP 23(3/4); 1993.

International Commission on Radiological Protection (ICRP). Human respiratory tract model for radiological protection. Oxford: Pergamon Press; ICRP Publication 66; Ann ICRP 24(1-3); 1994.

International Commission on Radiological Protection (ICRP). Individual monitoring for internal exposure of workers, replacement of ICRP Publication 54. Oxford: Pergamon Press; ICRP Publication 78; Ann ICRP 27(3/4); 1997.

Khokhryakov VF, Suslova KG, Kudryavtseva TI, Schadilov AE, Vostrotin VV, Lagounova NY, Barabanshchikova AY. Relative role of plutonium excretion with urine and feces from human body. Health Phys 86(5):523-527; 2004.

Khokhryakov VF, Suslova KG, Vostrotin VV, Romanov SA, Eckerman KF, Krahenbuhl MP, Miller SC. Adaptation of the ICRP publication 66 respiratory tract model to data on plutonium biokinetics for Mayak workers. Health Phys 88(2):125-132; 2005.

Knoll GF. Radiation detection and measurement. 3rd ed. New York: John Wiley & Sons, Inc.; 2000: 91-92.

Leggett RW. A retention-excretion model for americium in humans. Health Phys 62(4):288-310; 1992.

Luciani A, Polig E. Verification and modification of the ICRP-67 model for plutonium dose calculation. Health Phys 78(3): 303-310; 2000.

Luciani A, Doerfel H, Polig E. Sensitivity analysis of the urinary excretion of plutonium. Radiat Protect Dosim 93(2):179-183; 2001.

Marsh JW, Bailey MR, Birchall A. A step-by-step procedure to aid the assessment of intake and doses from measurement data. Radiat Protect Dosim 114(4):491-508; 2005.

Potter CA. Intake retention fractions developed from models used in the determination of dose coefficients developed for ICRP publication 68-particulate inhalation. Health Phys 83(5): 594-789; 2002.

Skrable KW, Chabot G, Harris J, French C. Dosimetric model for the gastrointestinal tract. Health Phys 28: 411-427; 1975.

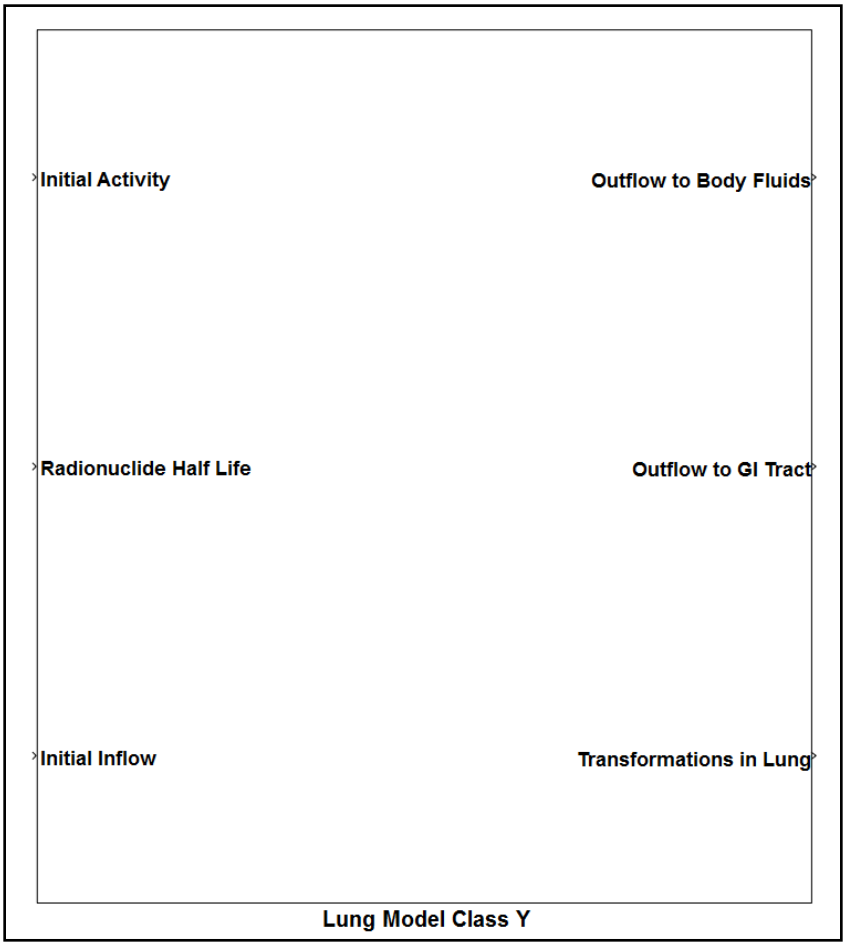
Task group on lung dynamics. Deposition and retention models for internal dosimetry of the human respiratory tract. Health Phys 12: 173-207; 1966.

Tuli JK, Burrows TW. Nuclear decay data in the MIRD format [online]. Available at: http://www.nndc.bnl.gov/useroutput/239pu_mird.html. Accessed 1 June 2007.

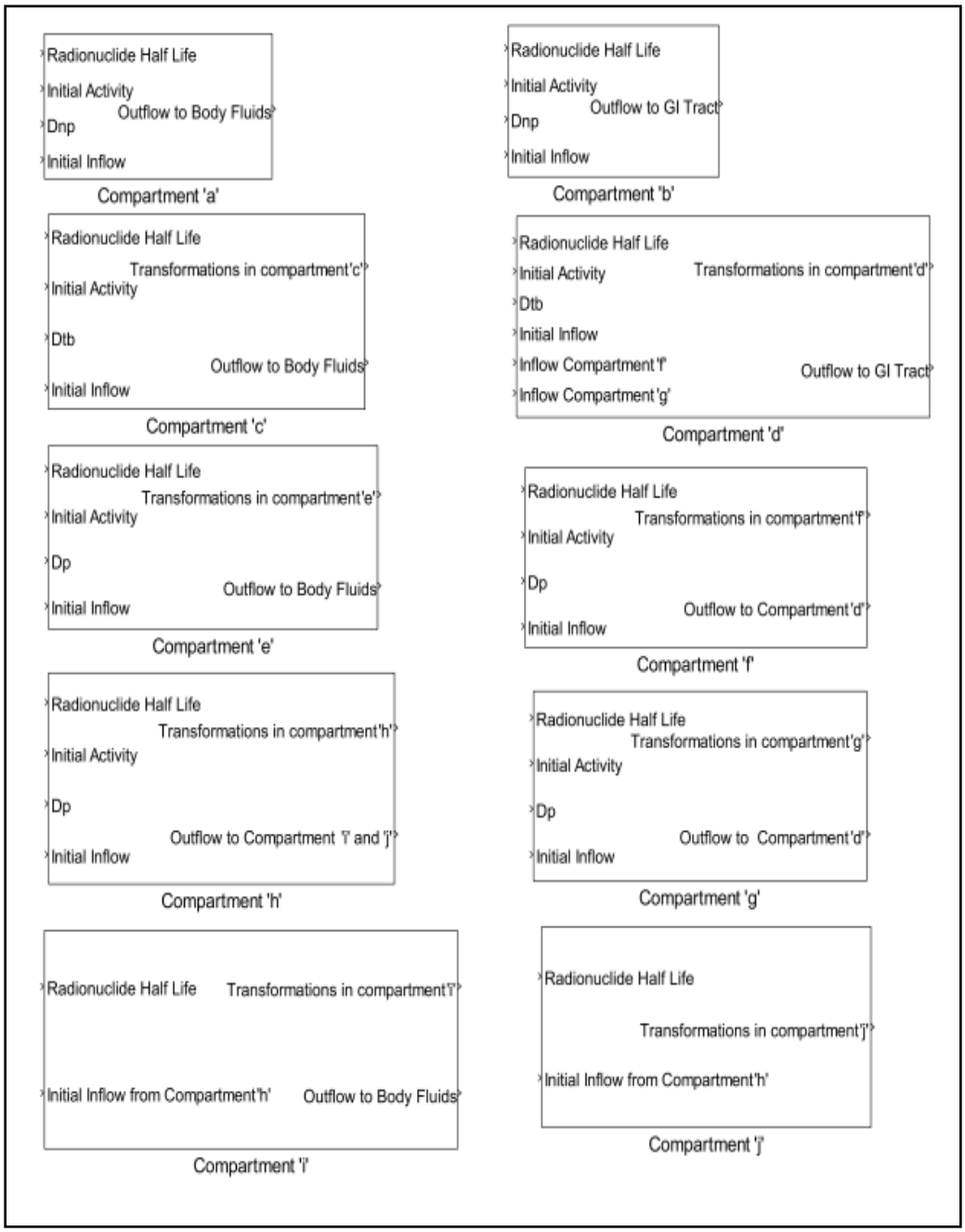
U.S. Nuclear Regulatory Commission. Acceptable concepts, models, equations, and assumptions for a bioassay program. Washington, DC: U.S. Nuclear Regulatory Commission; Regulatory Guide 8.9; 1993.

APPENDIX

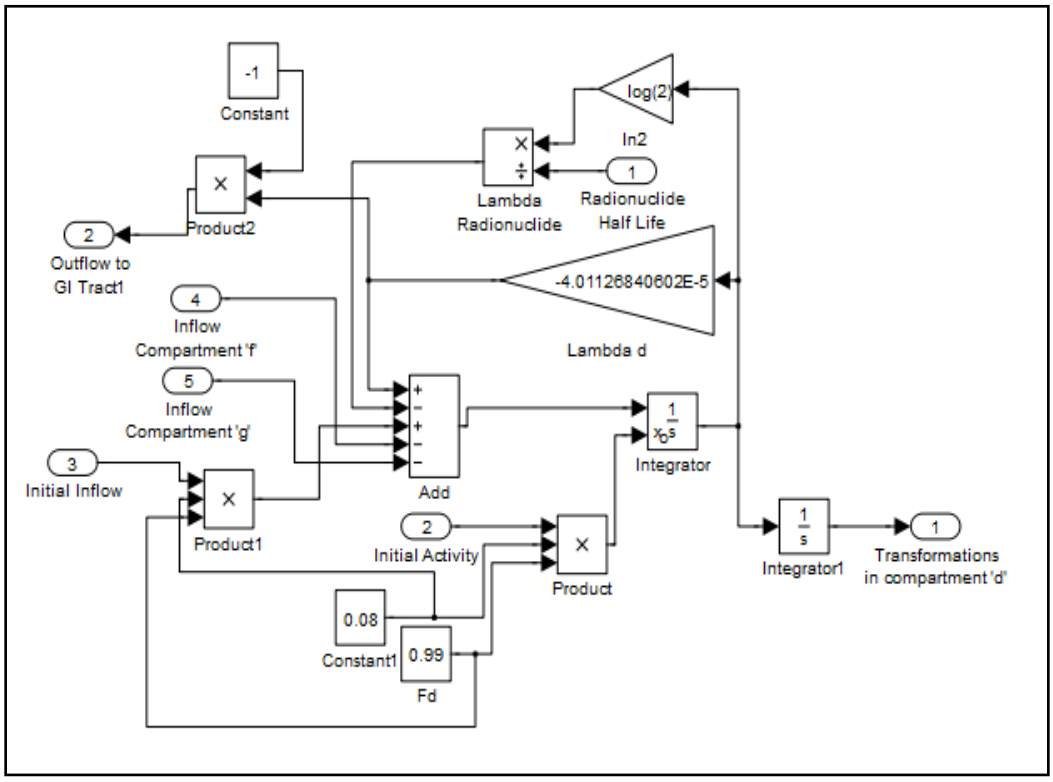
ICRP-30 RESPIRATORY TRACT MODEL



Upper level of ICRP-30 respiratory tract model

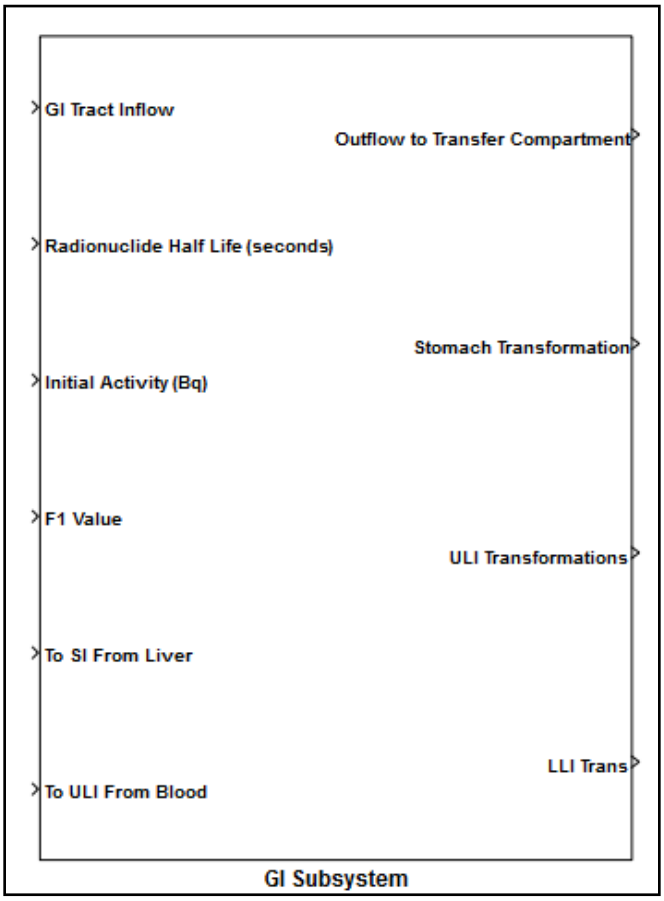


Middle level of ICRP-30 respiratory tract model



Lower level of ICRP-30 respiratory tract model

ICRP-30 GI-TRACT MODEL



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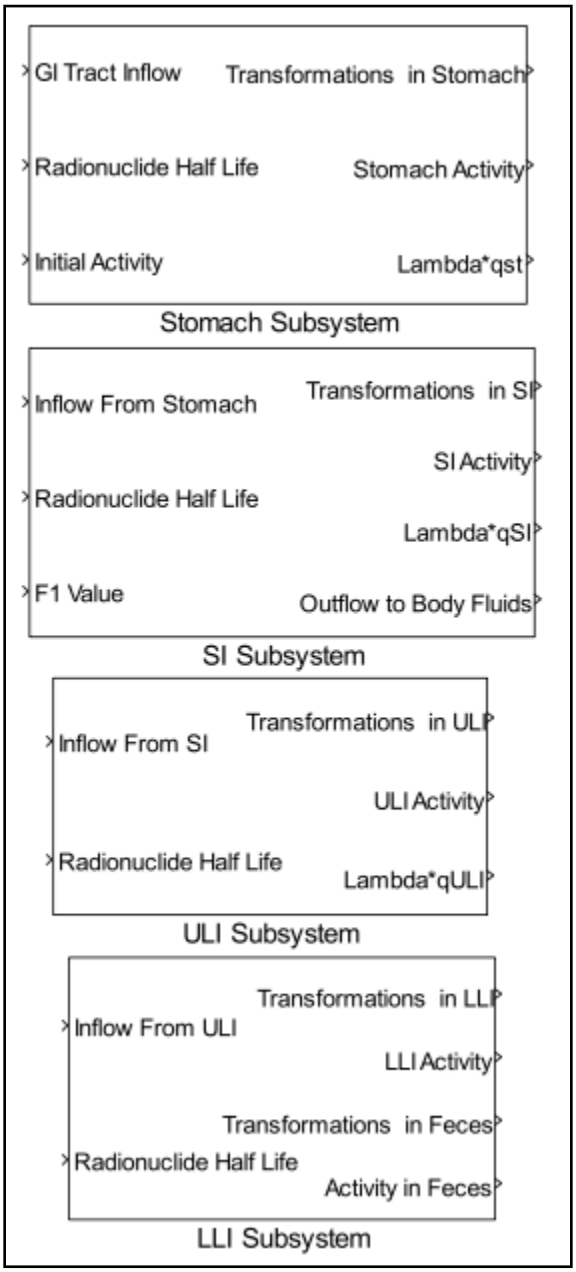

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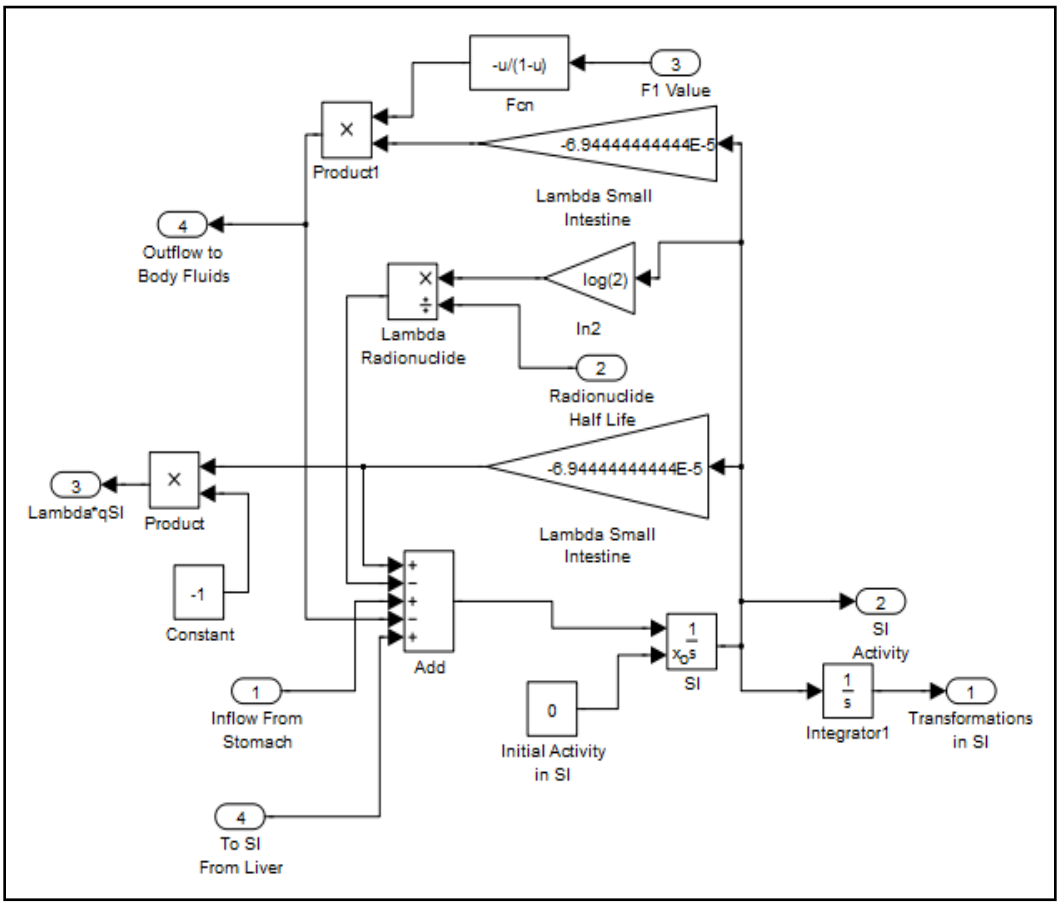
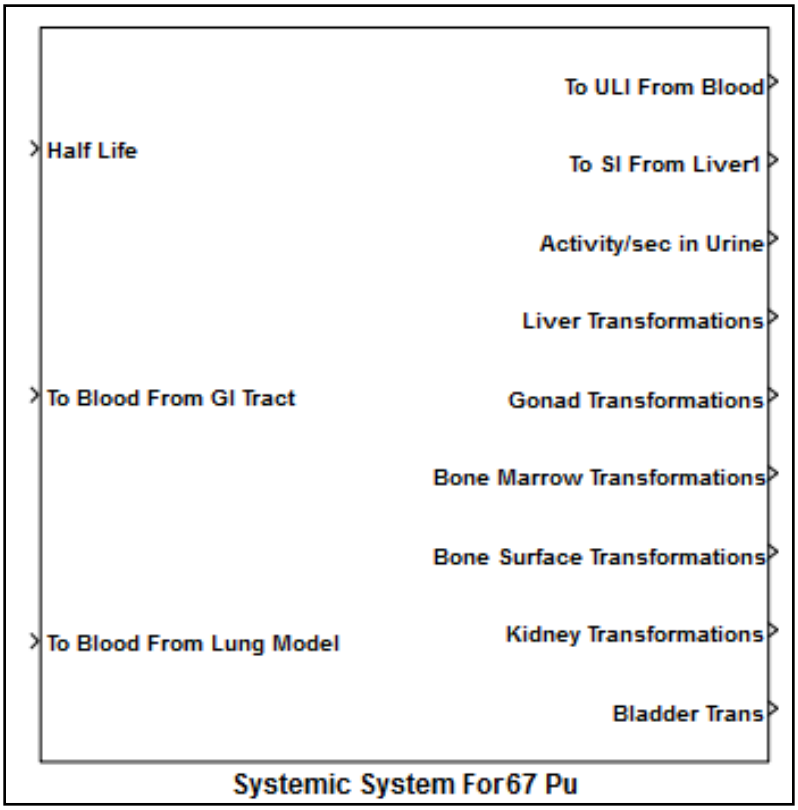
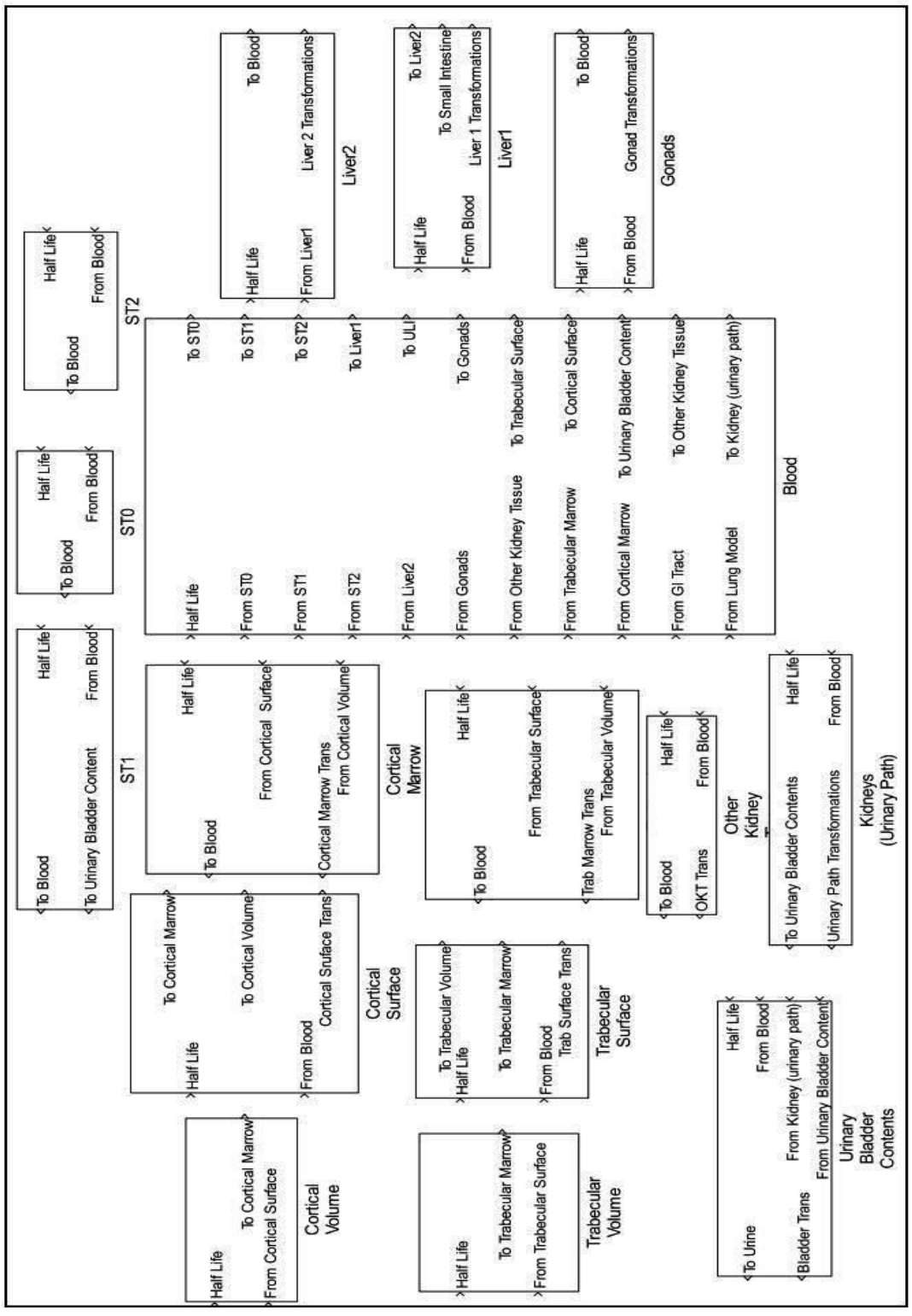


Lower level of the ICRP-30 GI-tract model

ICRP-67 PLUTONIUM SYSTEMIC MODEL



Upper level of the ICRP-67 plutonium systemic mod



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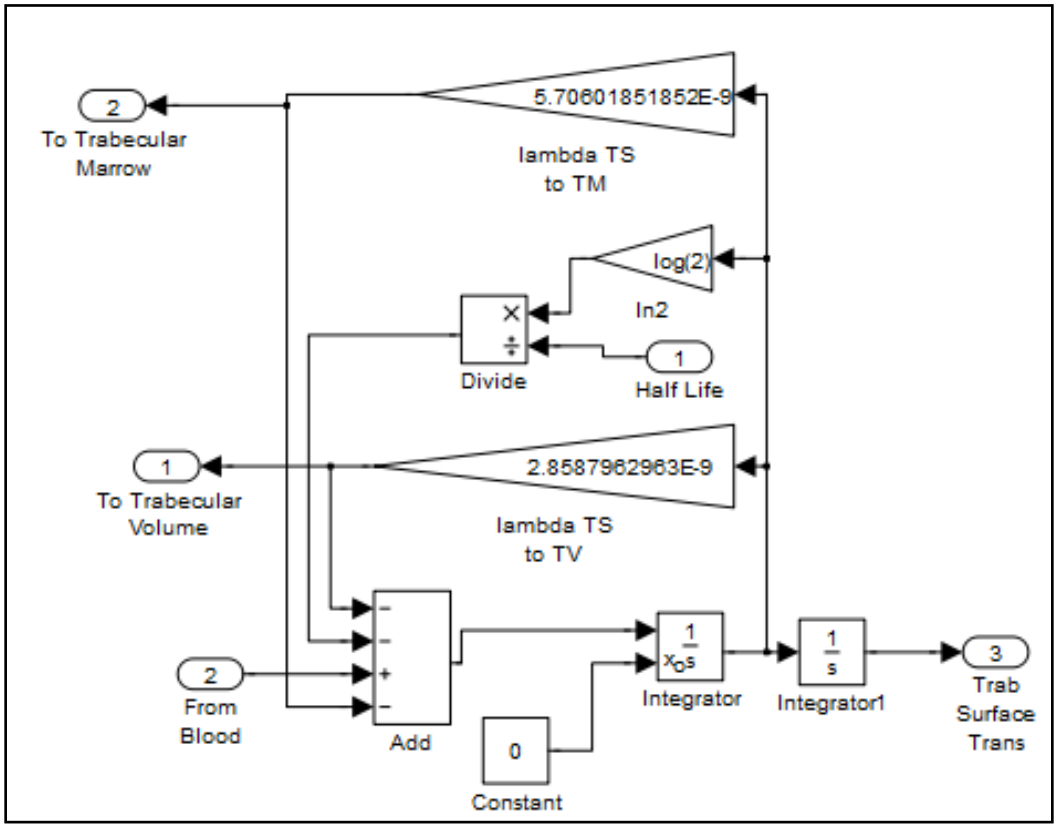
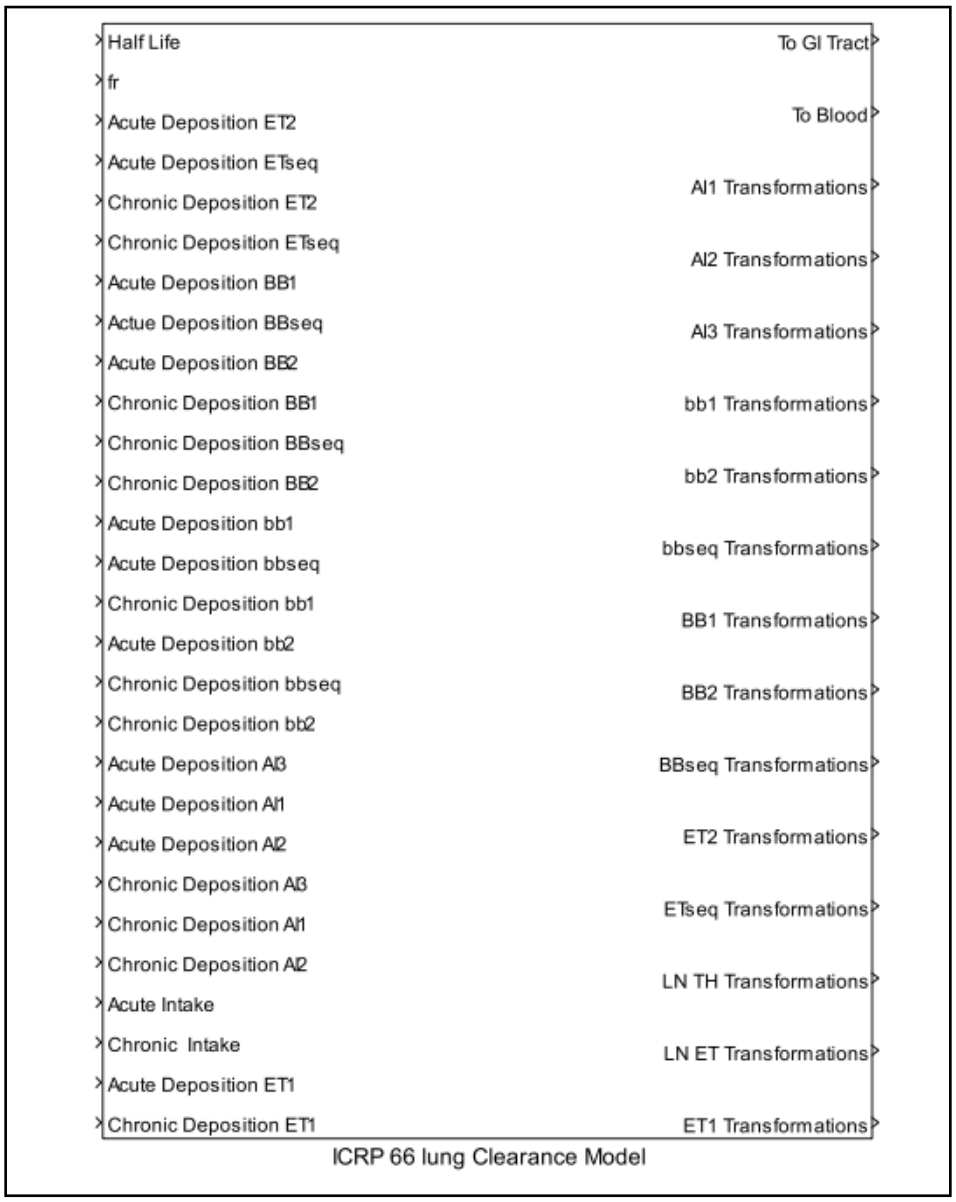


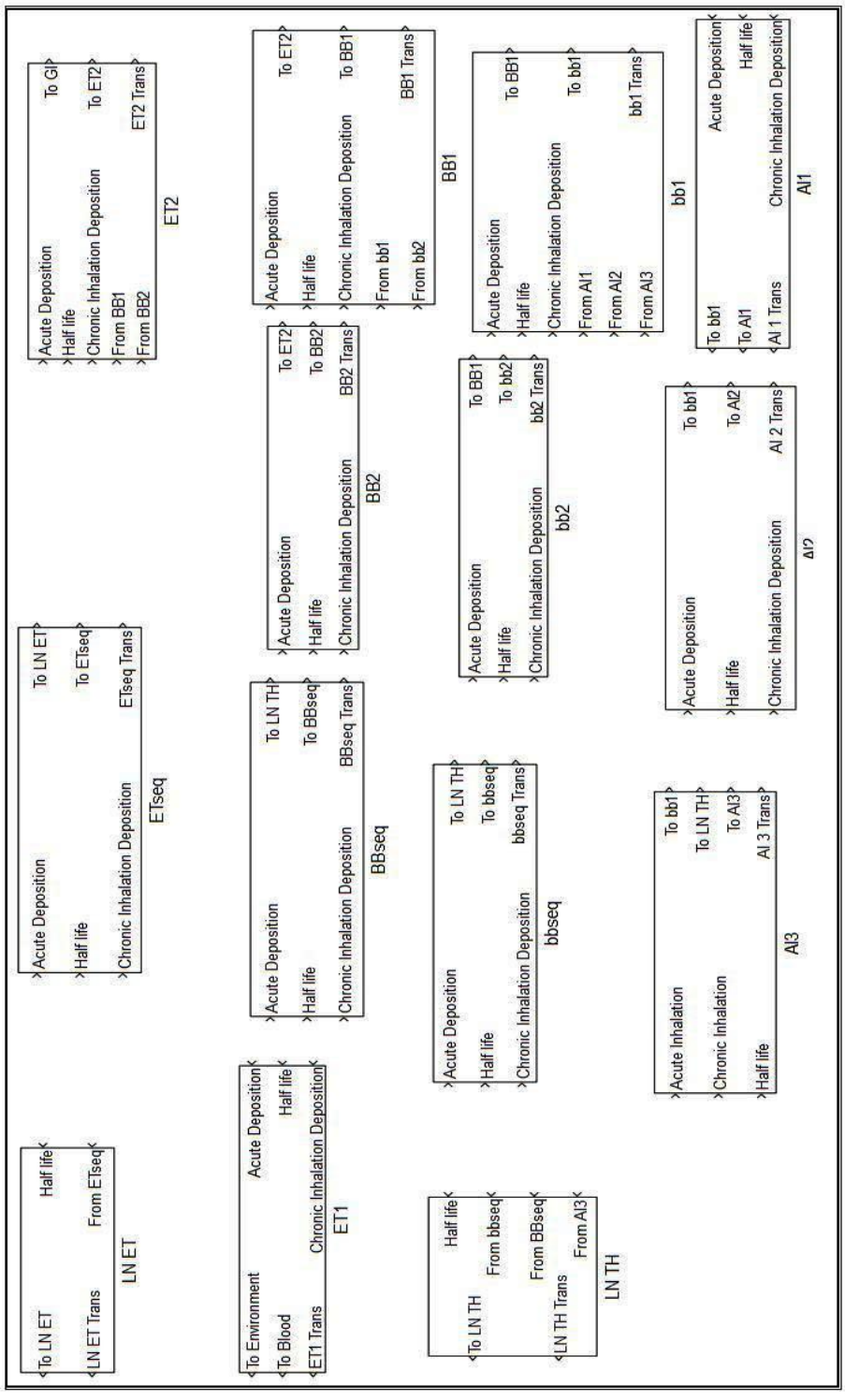
Lower level for the ICRP-67 plutonium systemic model

ICRP-66 RESPIRATORY TRACT MODEL

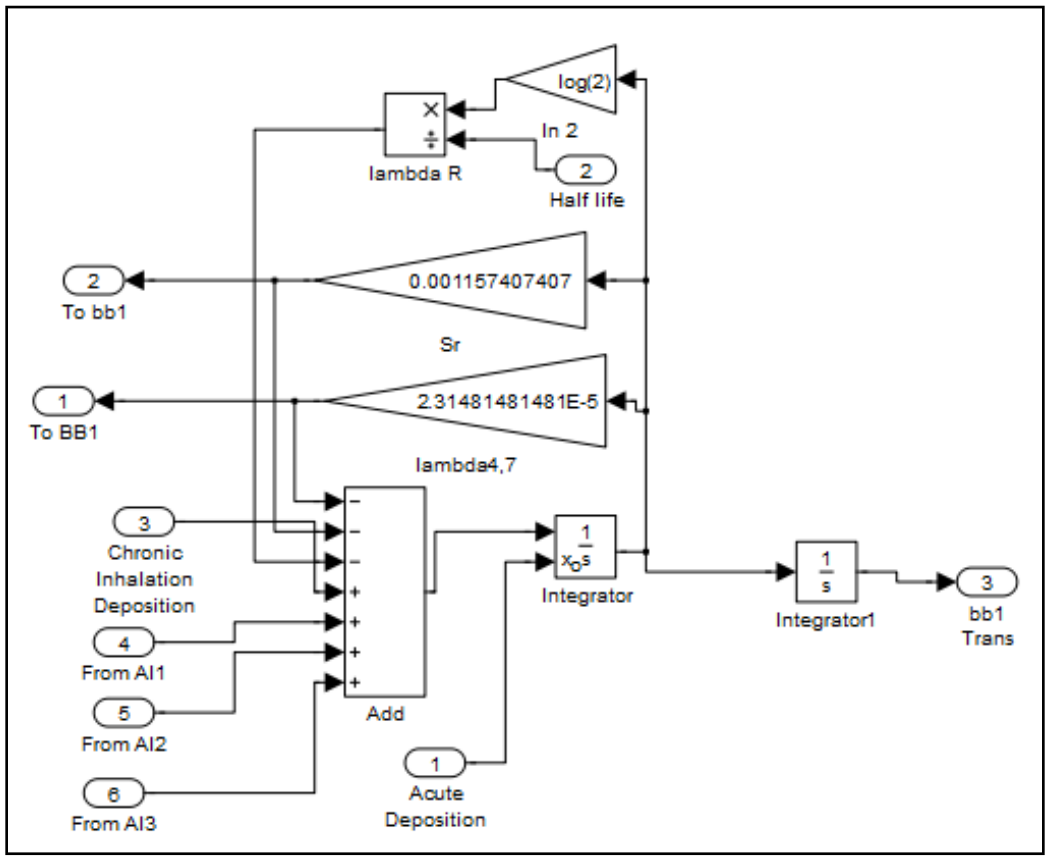


Upper level for the ICRP-66 respiratory tract model

Middle level of ICRP-66 respiratory tract model showing rapid and slow dissolution sections

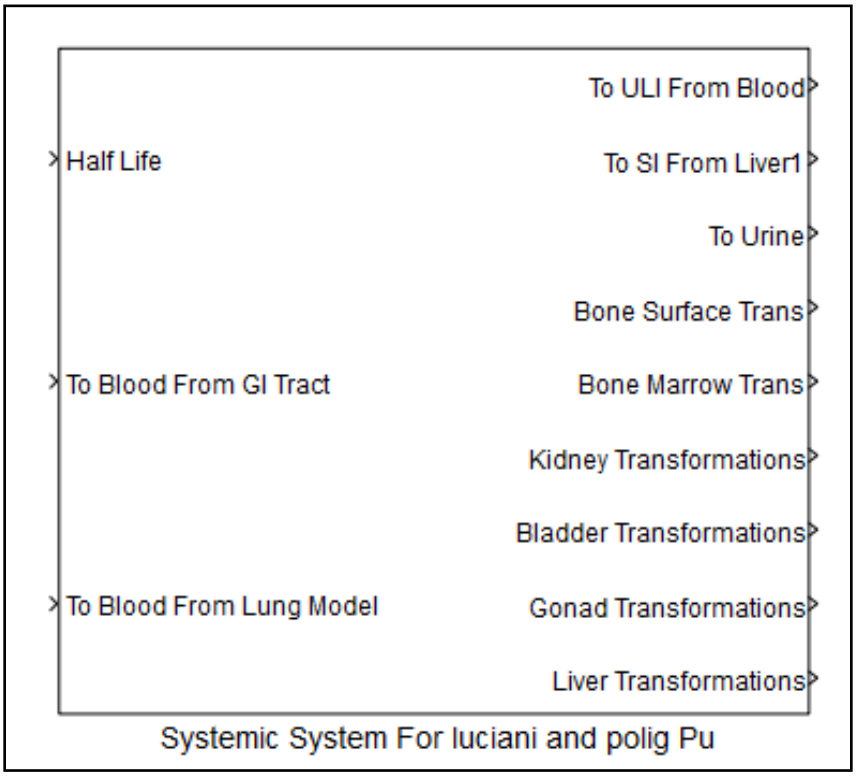


Middle level of the rapid dissolution section of the ICRP-66 respiratory tract model

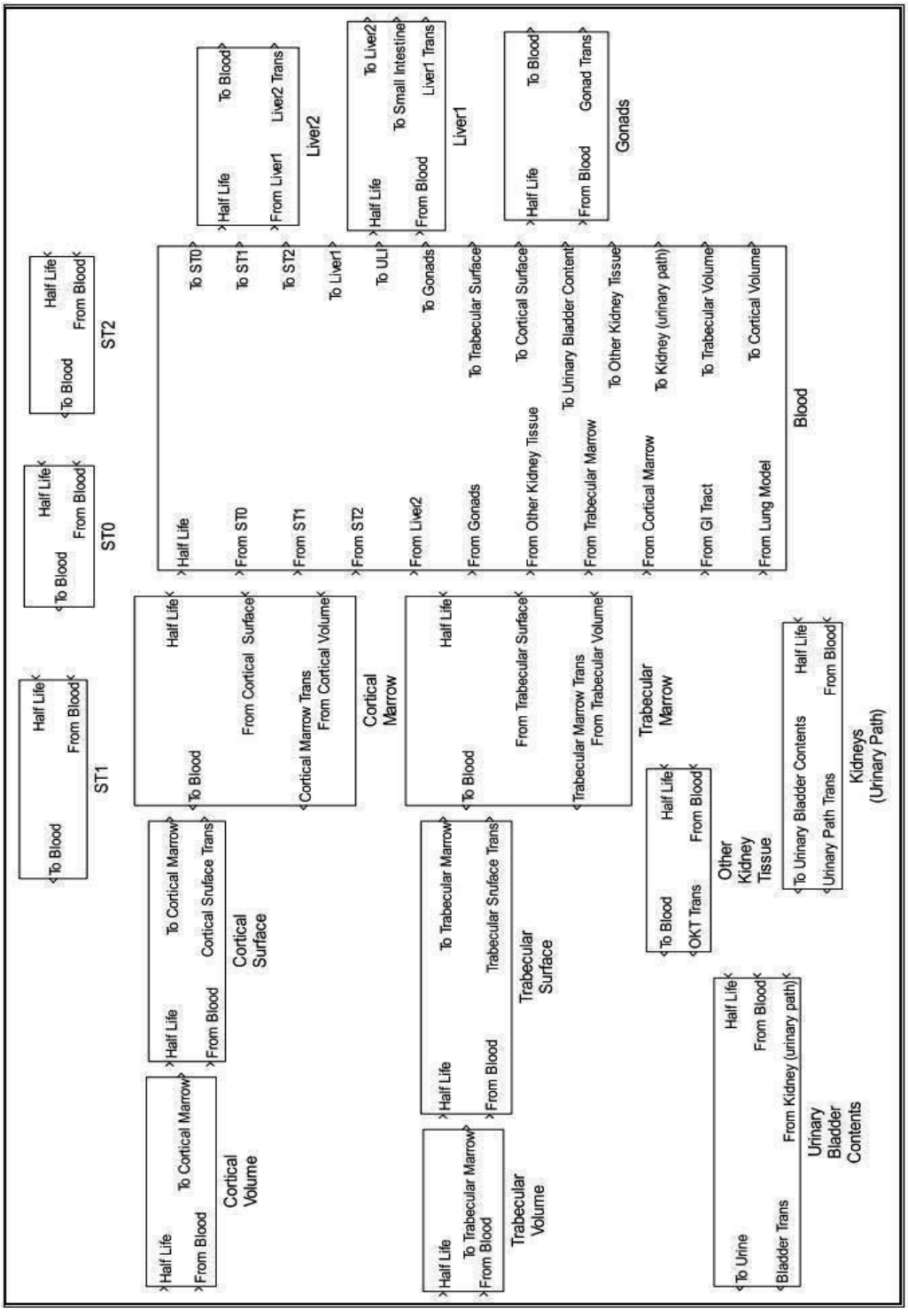


Lower level of the ICRP-66 respiratory tract model

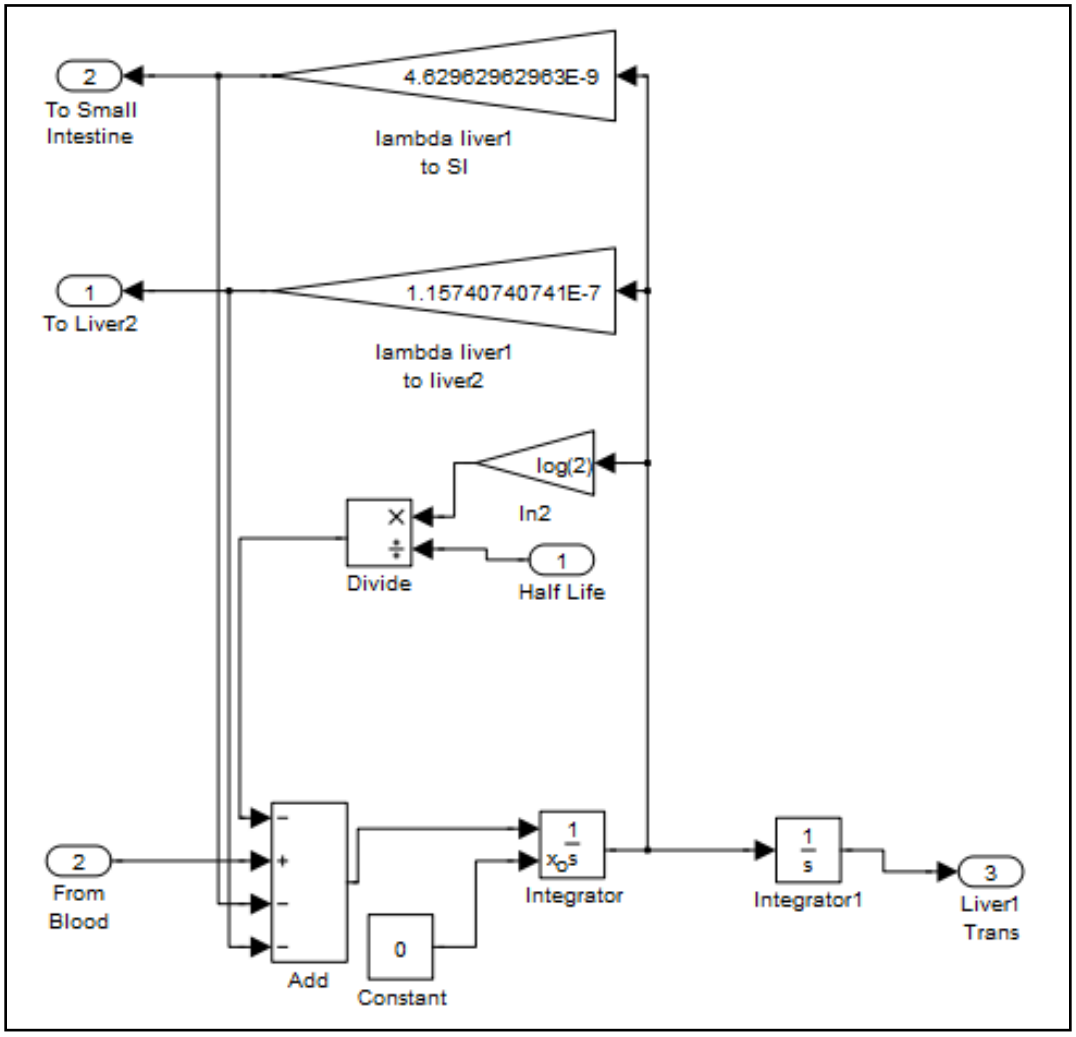
LUCIANI AND POLIG PLUTONIUM SYSTEMIC MODEL



Upper level of the Luciani and Polig plutonium systemic mode

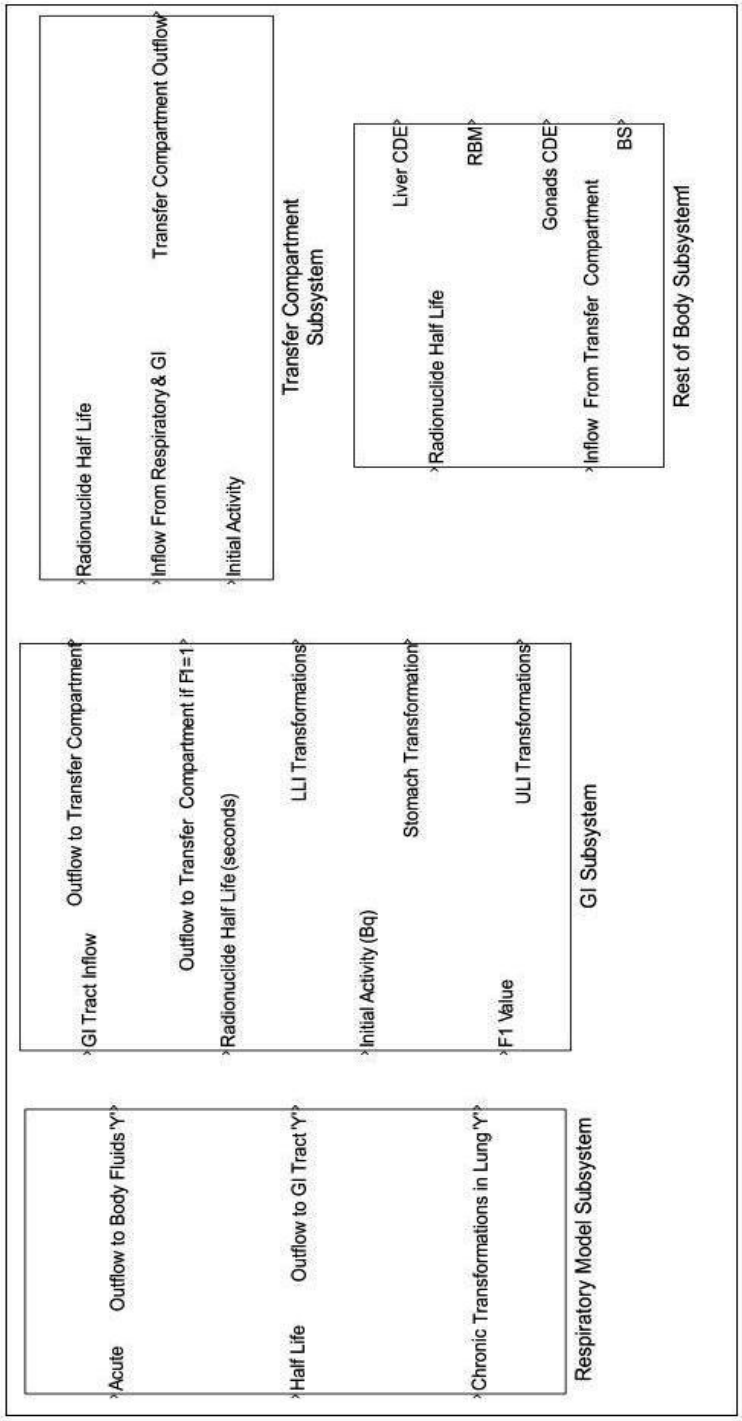


Middle level of the Luciani and Polig plutonium systemic model

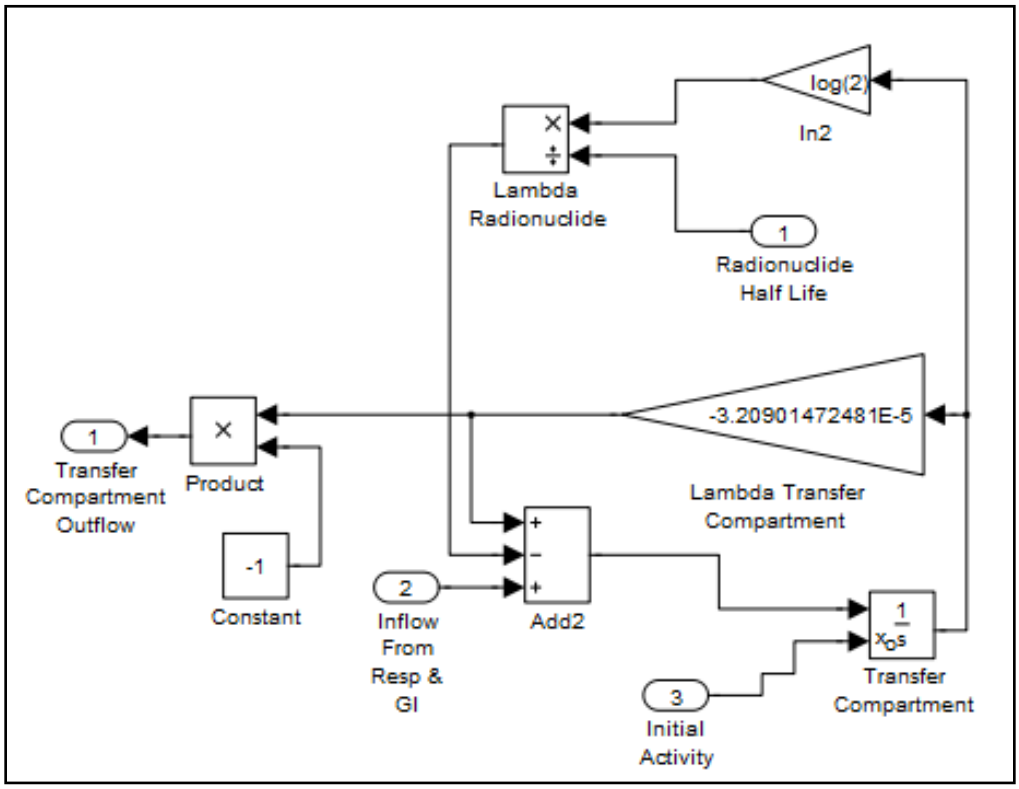


Lower level of the Luciani and Polig plutonium systemic model

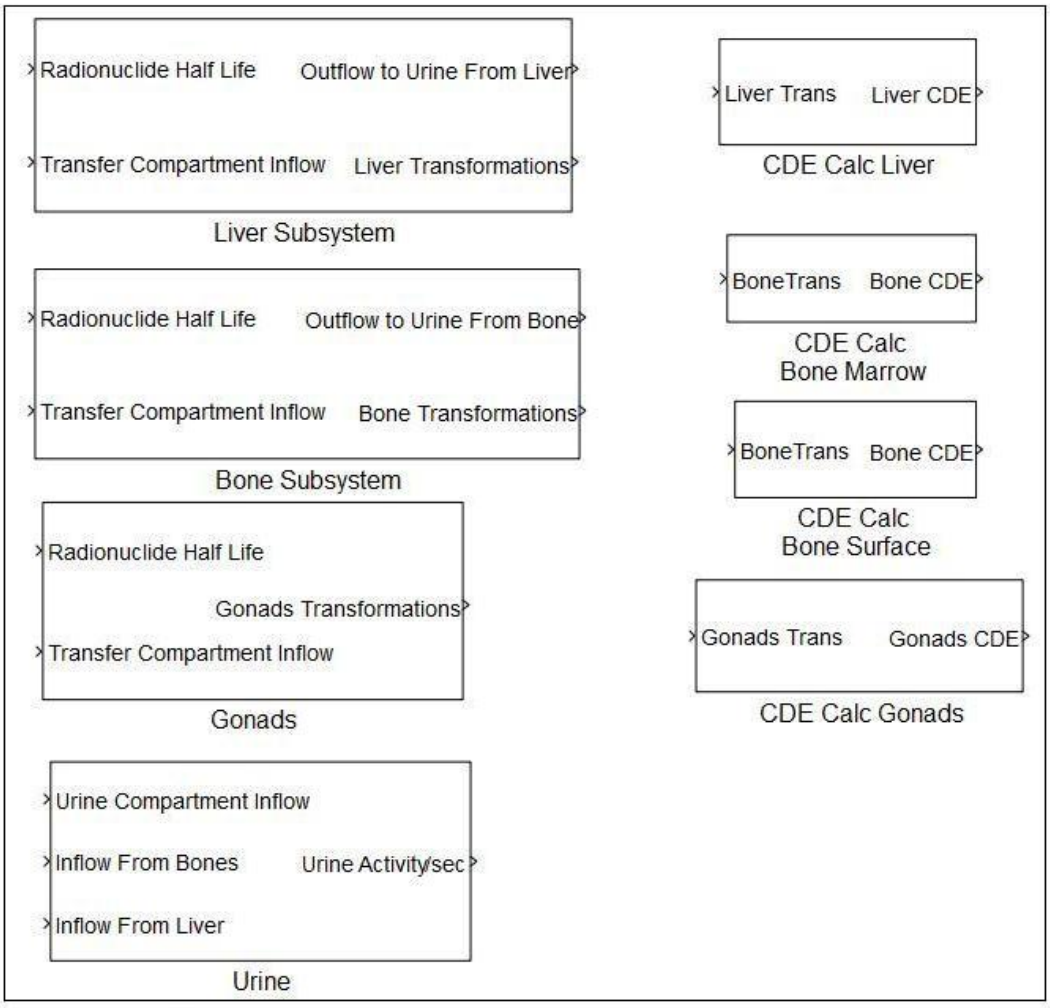
ICRP-30 DOSIMETRY MODEL



Combination of models that form the ICRP-30 dosimetry model



Lower level of the ICRP-30 dosimetry transfer compartment



Middle level of some of the main organs of interest in the ICRP-30 dosimetry model

VITA

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